

SHONKA RESEARCH ASSOCIATES, INC.

CALC NO SRA-96-010 REV 0

Project/Task ChemRisk/TDH Oak Ridge Dose Reconstruction

Prepared by R. E. Burmeister Date 10/31/96 Checked by/Date

Title Fitting Uranium Releases for ESA, EIVA, CIVA, and DD Pathways

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J. J. Shonka

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Calculation Control Sheet

Calculation number: SRA-96-010 REV. 0

Title: Fitting Uranium Releases for ESA, EIVA, CIVA, and DD Pathways

Reason for calculation/revision: New calculation

Client: ChemRisk/TDH

Project: Oak Ridge Dose Reconstruction

Project/Task Number: Task 6

Prepared by: Regan E. Burmeister
Regan E. Burmeister (signed original on file)

Date: 10/31/96

Independent Technical Review by: Joseph J. Shonka
Joseph J. Shonka (signed original on file)

Date: 10/31/96

Quality Assurance Review by: Deborah B. Shonka
Deborah B. Shonka (signed original on file)

Date: 10/31/96

☐ This calculation has been voided or superseded by

(calculation number)

#3189

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JS / 10/31/96
J.J. Shonka
(signed original on file)Title Fitting Uranium Releases for ESA, EIVA, CIVA, and DD Pathways**Review Method Sheet**

The undersigned has reviewed this calculation in accordance with the method(s) indicated below.

1. Computer Aided Calculation	
a	Review to determine that the computer program(s) has been validated and documented, is suitable to the problem being analyzed, and that the calculation contains all necessary information for reconstruction at a later date.
b	Review to determine that the input data as specified for program execution is consistent with the design input, correctly defines the problem for the computer algorithm and is sufficiently accurate to produce results within any numerical limitations of the program.
c	Review to verify that the results obtained from the program are correct and within stated assumptions and limitations of the program and are consistent with the input.
d	Review validation documentation for temporary changes to listed, or developmental, or unique single application programs, to assure that the methods used adequately validate the program for the intended application.
e	Review of code input only, since the computer program has sufficient history of use at Shonka Research Associates, Inc. in similar calculations.
f	Review arithmetic necessary to prepare code input data.
g	Other:
2. Hand Prepared Calculations	
a	Detailed review of the original calculations.
b	Review by an alternate, simplified, or approximate method of calculation.
c	Review of a representative sample of repetitive calculations.
d	Review of the calculation against a similar calculation previously performed.
e	Other:
3. Revisions	
a	Editorial changes only
b	Elimination of unapproved input data without altering calculated results.
c	Other:
4. Other	

Reviewer:

Joseph J. Shonka (signed original on file)

Date:

10/31/96

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ABSTRACT

This calculation presents work done to estimate the uranium release from the K-1131, K-1401, and K-1420 buildings through the ESA pathway and from the various diffusion cascade buildings through the EIVA, CIVA, and DD pathways. Release data existed for these buildings and pathways for particular time periods but was incomplete; it was known that the time periods were longer, but data was unavailable or did not exist in order to complete the release histories for the buildings. To estimate the unknown release amounts, statistical analyses were performed on the existing data. Governing probability distributions with their concomitant means, mean standard errors, and percent certainty ranges were determined. This information allowed statements to be made about the release amounts for the unknown time periods.

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1. INTRODUCTION

Task 6 of the Oak Ridge Dose Reconstruction focuses on the evaluation of the quality of historical airborne and waterborne effluent monitoring data and the determination of the potential significance of unmonitored emissions. Uranium played an important role throughout historical operations on the Oak Ridge Reservation (ORR) and is known to have been released to the environment through air and water. The two largest uses of uranium on the Reservation were the enrichment processes of the ^{235}U isotope by electromagnetic separation at the Y-12 facility and gaseous diffusion at the K-25 facility.

For convenience, Task 6 of the Oak Ridge Dose Reconstruction divided potential releases from diffusion cascade into four release pathways. The ESA pathway began with process equipment that led to evacuation and exhaust equipment that finally led to the atmosphere. The EIVA pathway began with process equipment that led to circulation in indoor air which led to building ventilation that led finally to the atmosphere. The CIVA pathway was the same as the EIVA pathway except that it began with cylinders that held feed, product, or tail material. The DD pathway began with tanks or drums that led to drain equipment. The tanks or drums were stored in the vaults located in the basement areas of the purge cascade.

The purge cascade statistical analysis for the ESA pathway was completed in a different calculation (SRA-96-009). The K-1131, K-1401, and K-1420 buildings also had ESA pathways for uranium release. The K-1131 building was primarily involved in UF_6 feed manufacturing, feed vaporization, and tails withdrawal. From 1951 to 1961 K-1131 was the production facility for the conversion of UF_4 to UF_6 . The conversion process underwent many design changes and contributed many large releases. The facility eventually managed to produce 18 tons per day of feed product. In March of 1962 the facility was reactivated to produce UF_6 from slightly enriched UF_4 for a three year period. The facility was finally deactivated in July 1965.

The feed vaporization facility of K-1131 heated cylinders of UF_6 feed to convert the solid UF_6 to liquid and vapor phases. During heating, the greatest potential and highest rate of release existed when the liquid UF_6 had a vapor pressure greater than atmospheric pressure. Such a situation was part of the vaporization process at K-1131. The feed vaporization facility operated from 1965 to 1985.

The tails withdrawal facility at K-1131 operated from 1962 to 1984. Its purpose was to remove depleted UF_6 from the enrichment cascade. The vapor from the cascade was converted to a liquid phase and then drained into containers. During this process, the liquid had a vapor pressure greater than atmospheric pressure, and this again created a situation for the greatest potential and highest rate of release.

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The K-1401 building operated as a decontamination and recovery facility from 1944 to 1985. Its primary purpose was the cleaning, conditioning, and assembly of equipment.

The K-1420 building was another decontamination and recovery facility which began operation in 1953. From 1954 to 1960, a uranium oxide fluorination process was operated in the facility. Emissions of UO_2F_2 and HF from the conversion reactors were vented directly to a stack. Caustic scrubbers were not installed until later and served to reduce UO_2F_2 emissions.

Tanks or drums of uranium solution were stored in the basement vaults of the cascade buildings and K-1131 throughout the operating history of the cascade. Corrosion of the drums was by far the dominant cause of leaks and by far the dominant contributor to events in the DD pathway.

The cascade buildings were as follows: K-25, K-27, K-29, K-31, and K-33. Each of these buildings were divided into cells which housed a number of diffusion stages. During the performance of maintenance activities, UF_6 had to be evacuated from sections of the cascade, and the system was opened to the atmosphere. Ambient moist air consequently contaminated the equipment. After the system was closed and leak checked, the wet air was ejected through jets and pumps to the atmosphere. The systems evacuated ranged from a few cubic feet in volume to several thousand cubic feet; wet air evacuation could contain significant amounts of uranium.

Compressors were used throughout the K-25 site for pumping UF_6 , and they were equipped with special shaft seals. Each of these seals was connected to an exhaust system designed to discharge exhaust gases from the seals to the atmosphere. Seal exhaust systems consisted of chemical traps for UF_6 , pumps, control instrumentation, connecting piping, and discharge stacks. The failure of a seal always created the probability of UF_6 being evacuated through the seal into the exhaust system and then to the atmosphere.

The K-25 building consisted of the following cascade cells: K-301-1 to K-301-5, K-302-1 to K-302-5, K-303-1 to K-303-10, K-304-1 to K-304-5, K-305-1 to K-305-10, K-306-1 to K-306-7, K-309-1 to K-309-3, K-310-1 to K-310-3, K-311-1, and K-312-1 to K-312-3. K-25 began operations in January 1945 and ceased gaseous diffusion operations in June 1964. In addition to the seal exhausts and wet air venting common to all cascade buildings, the K-312-1 to K-312-3 and K-311-1 cells operated as the purge cascade unit for much of the history of the K-25 plant. Purge cascade emissions were estimated and documented in SRA-95-002, 011, 012, 013.

The K-27 plant consisted of the cascade cells K-402-1 to K-402-9 and began operations in December 1945. Operations ceased in June 1945. Seal exhausts and wet air evacuation were common occurrences.

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K-29 began operations in September 1950 and ceased operation in 1985. Only three cells K-502-1 to K-502-3 made up the K-29 building, but one wet air evacuation system located in cell K-502-2 served all three cells.

K-31 began operation in August 1951 and ceased operation in 1985. The cells K-602-1 to K-602-6 made up the K-31 building. Seal exhausts and wet air evacuation also occurred in K-31.

K-33 began operation in March 1954 and ceased operation in 1985. The cells K-901-1 to K-901-8 made up the K-33 building. Seal exhausts and wet air evacuation also occurred in K-33.

Equipment → Indoor → Ventilation → Atmosphere
For the pathways EIVA, CIVA, and DD, all the cascade buildings and cells were grouped under the general class 'cascade', as distinct from the 'purge' cascade studied in SRA-96-009. The releases from the cascade through the EIVA, CIVA, and DD pathways and the releases for buildings K-1131, K-1401, and K-1420 through the ESA pathway in addition to releases for K-1131 for EIVA and CIVA pathways were retrieved from site records and entered into the ORHS-II Master Release List (SRA-96-012). These releases were statistically analyzed to determine the probability distributions that best described the release data for a particular building and pathway. The distributions were then sampled in order to make a bounded estimate of release which was then used to fill in the gaps in the release history of these buildings.

K-1131, K-1401, K-1420, and the cascade were selected for distribution analysis because a preliminary analysis of releases in the ORHS-II Master Release List and a knowledge of the historical activities on the K-25 site led to the conclusion that releases from these buildings dominated other buildings for particular pathways. See Figures 1.1 - 1.8. which provide pie

**Distribution of Number of Uranium Releases
Category "EIVA"**

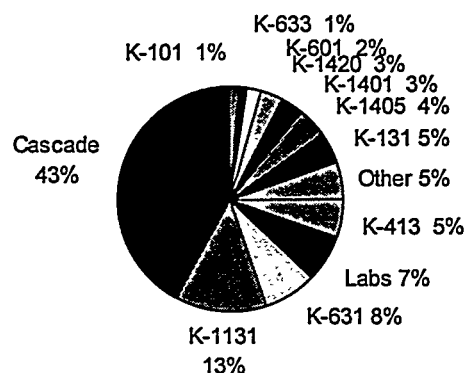


Figure 1.1 Distribution of Uranium Releases - Category EIVA

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charts showing either percentage total mass released or alternately percentage number of releases for each of the four release categories as a function of building.

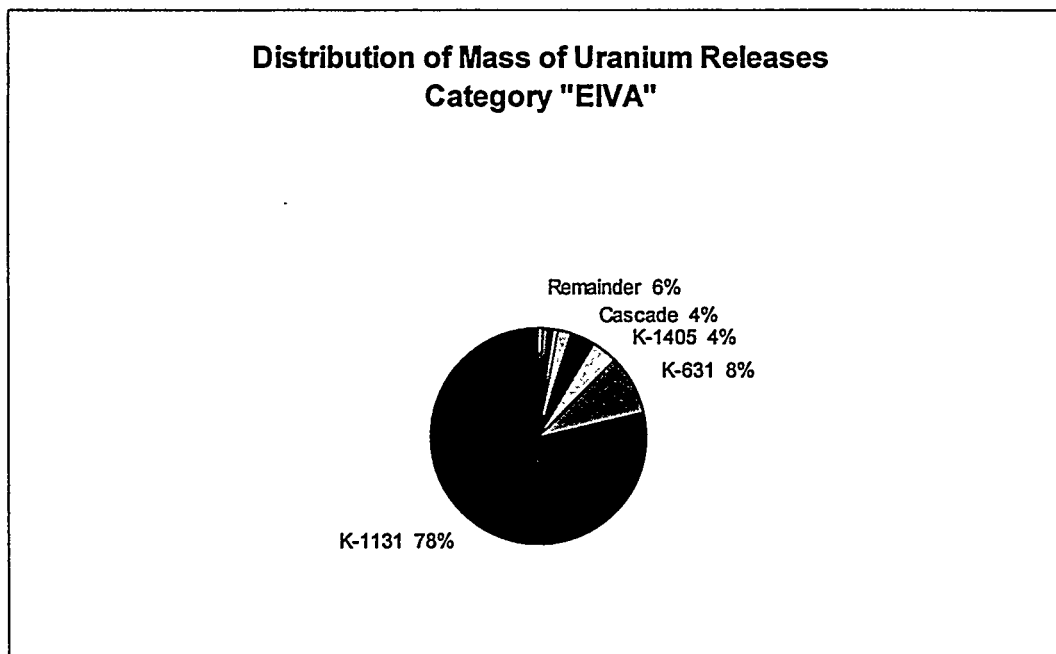


Figure 1.2 Distribution of Mass of Uranium Releases - Category "EIVA"

*Tall boxes on left wall
binders w/ photo from OK
picture men dumping mrc. into trough
picture of flask - thinks they are
separated slides from prints
Red EX to Nashville*

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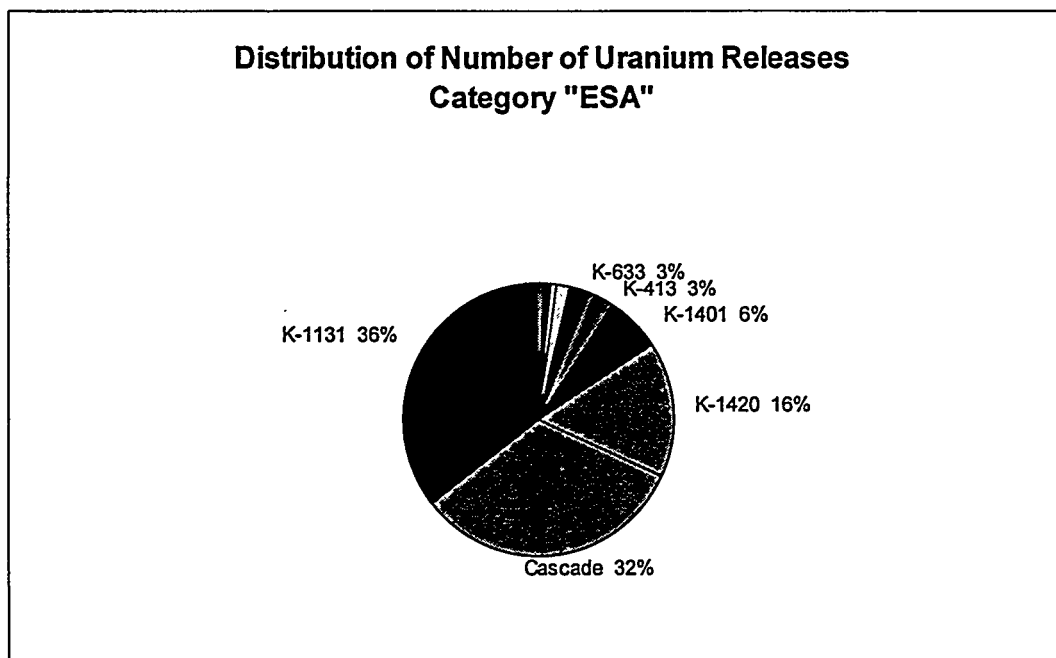


Figure 1.3 Distribution of Number of Uranium Releases - Category "ESA"

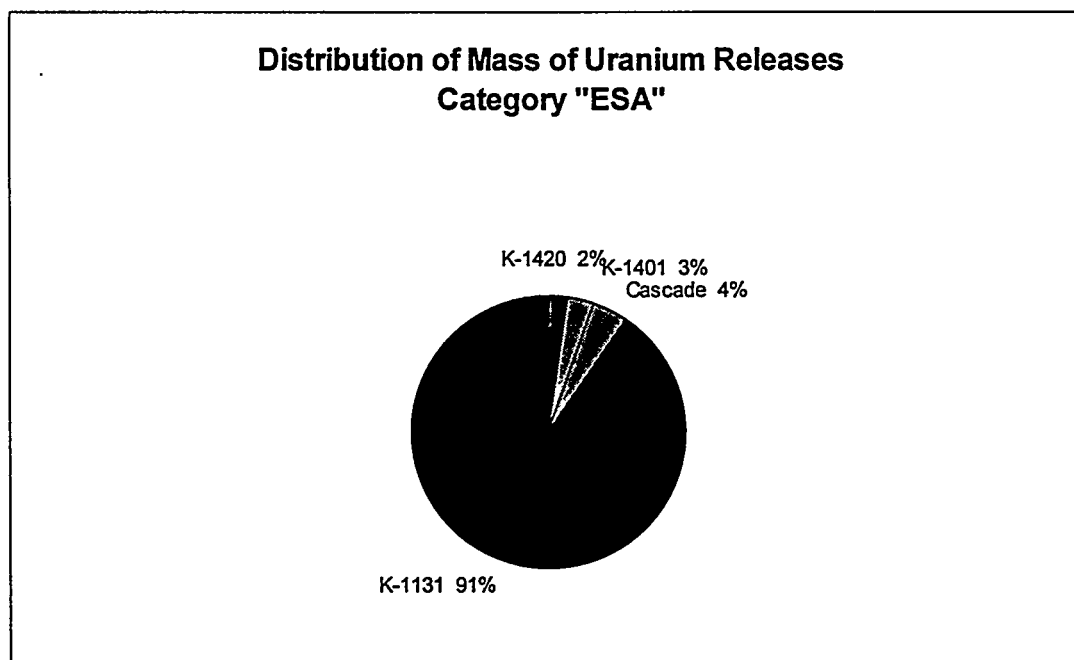


Figure 1.4 Distribution of Mass of Uranium Releases - Category "ESA"

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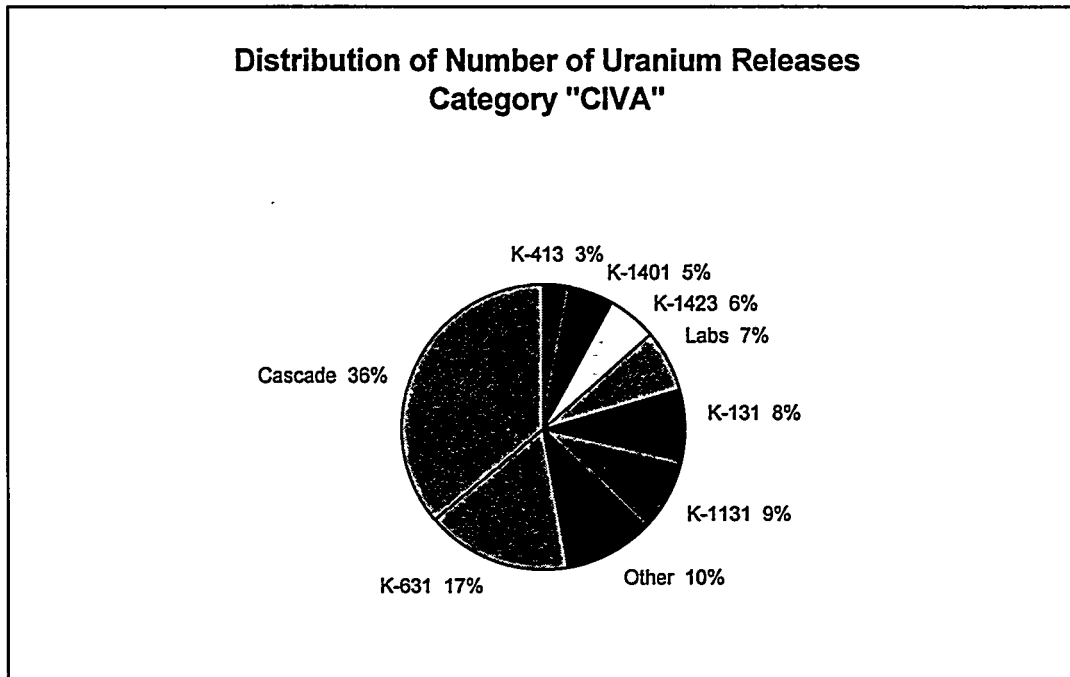


Figure 1.5 Distribution of Number of Uranium Releases - Category "CIVA"

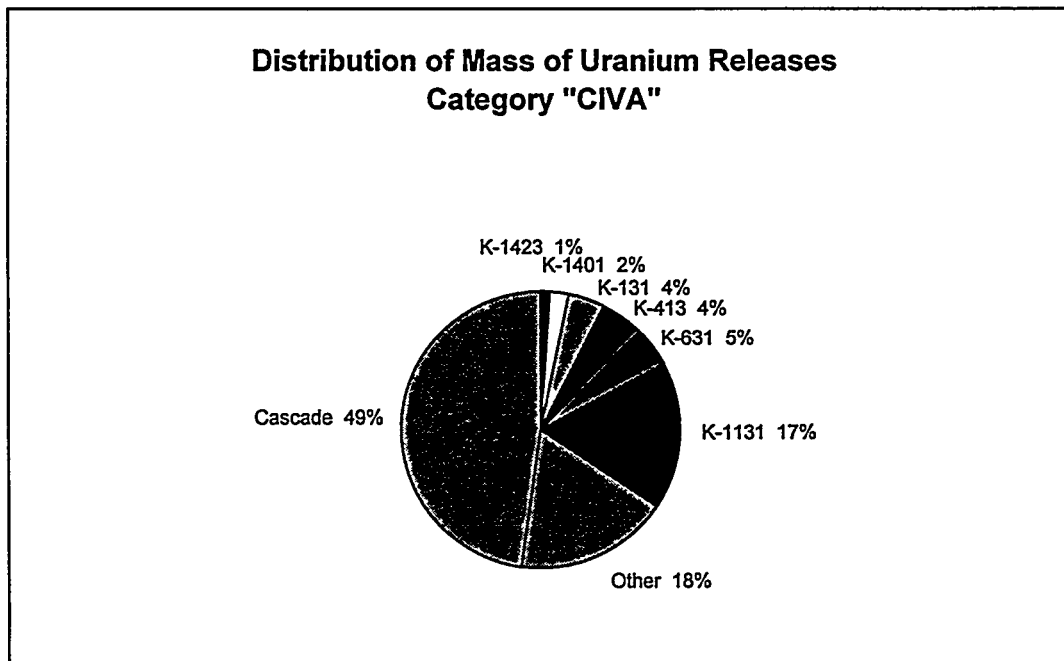


Figure 1.6 Distribution of Mass of Uranium Releases - Category "CIVA"

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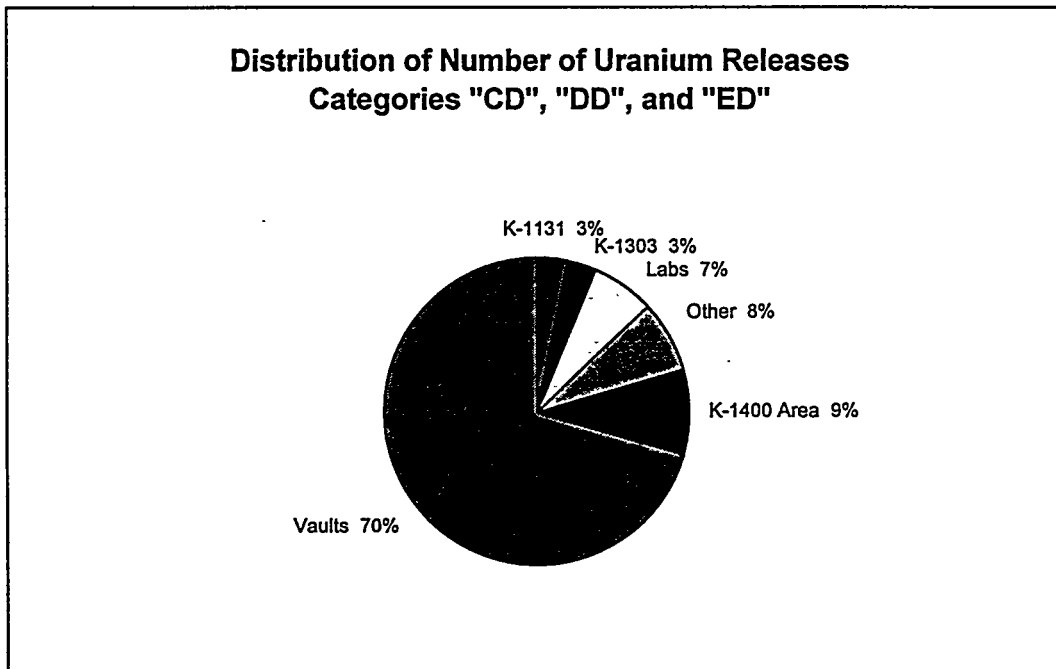


Figure 1.7 Distribution of Number of Uranium Releases - Categories "CD", "DD", & "ED"

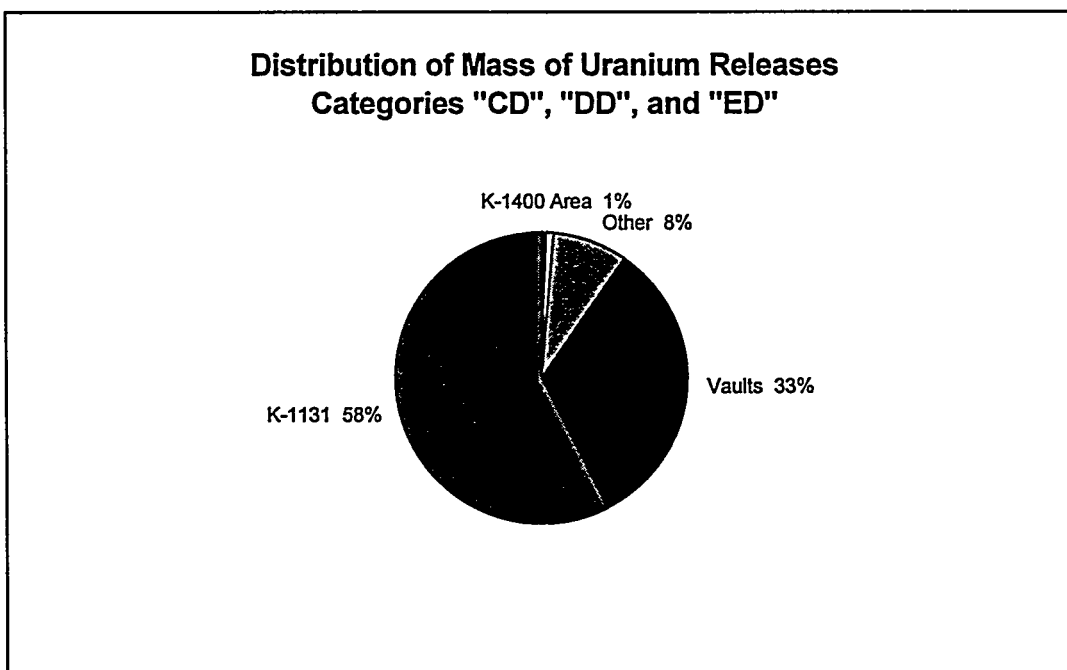


Figure 1.8 Distribution of Mass of Uranium Releases - Categories "CD", "DD", & "ED"


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2. SUMMARY OF RESULTS

The following table reports the K-1131, K-1401, and K-1420 fitted release amounts for the ESA pathway.

Table 2.1 Fitting Results for ESA Pathway for K-1131, K-1401, and K-1420

Building	Fitted Mass Release (g)	Lower 95 % Bound (g)	Upper 95% Bound (g)
K-1131	391735	262815	544044
K-1401	114332	98721	136421
K-1420	18787	5105	57061

Each building's release data was statistically analyzed to determine the probability distribution that best described the release data. Once the distribution and its descriptive parameters were known, an annual release could be simulated by assuming 12 monthly releases, each month having the determined probability distribution. In this way annual release amounts were estimated. This value is reported in the Fitted Mass Release column. The Lower 95% and Upper 95% Bounds are the 95% certainty bounds on the correction mass; i.e. the actual release has a 95% probability of lying between the bounds, if the distribution of releases was properly represented by the sample and if the asserted releases had no bias.

The cascade data for both the EIVA and CIVA pathways were very sparse. Although release incidents were recorded, the records did not indicate release amounts. For the EIVA pathway, the cascade dominated the number of release when compared to other buildings; 43% of the number of releases as shown in Figure 1.1., but the mass contribution is only 4% as shown in Figure 1.2. Upon examination of the data, it was found that only a few sporadic incidents had mass release values recorded; most of the data was just a record of when and what kind of incident had occurred. A distribution could have been fit to this data, but the fit would have been very poor. Furthermore, any predictive statements about mass releases based on such a fitted distribution would have had little confidence. In view of this situation, no fits were made for the cascade - EIVA pathway data.

The cascade - CIVA pathway data was also dominated by the cascade in terms of the number of releases as shown in Figure 1.5. The release data contained, however, very few records of release amounts, and of the few recorded amounts, there were some large releases. This situation explains the mass distribution shown in Figure 1.6 which does not indicate the sparseness of data and lack of mass values. As for the EIVA pathway, no fits were made for the CIVA pathway due to the sparse nature of the data set.

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The data sets for the K-1131 building for both the EIVA and CIVA pathways were also likewise very sparse. For the same reasons as above, no fits were made.

The determination of release estimates for the DD pathway involved two different fits to the available data. The number of releases or leaks per year, and the amount of material per release were both studied statistically to determine the probability distributions that best described the data. The two results were multiplied together to yield an annual release distribution. This annual release was 25,864 g of uranium with a 95% certainty between 444 g and 370,000 g of uranium. The annual release did not include any reductions due to drain filters or settling.

The annual release distribution was extremely positively skewed. That was the reason for the very large upper 95% bound, and in fact the mode of the distribution, i.e. the most probable value, rather than the mean was used for the annual release since the mean was more than a factor of two greater than the mode. Using the mean would have overstated the true magnitude of the release.

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3. METHODS

Two primary methods were used to generate the mass corrections. A commercially available software package for statistical analysis was first used to analyze a particular time period's data. This software determined the type of probability distribution that best described that data. A second commercially available software package for forecasting and risk analysis was used to generate a predicted release mass with upper and lower limits that reflected a 95% certainty in the release amount. The input to this software was the distribution type and parameters determined by the statistical analysis software.

The fitting was performed using the "Probability Distribution Plotting" software (PDP) (TEAM). Following examination of the PDP output, the appropriate distribution was selected, and its coefficients were entered into the modeling software, where the risk assessment was made by Crystal Ball (Crystal). Use of PDP was necessary because Crystal Ball did not provide fitting to distributions. A brief description of the software and their use is as follows:

Using PDP, the data was entered and computations were made for each of the following distributions: normal, lognormal, extreme value, log extreme value with both left and right skew, three parameter Weibull, and a gamma-corrected two-parameter Weibull. Both generalized least squares (GLS) and Ferrell's median regression (MRL) models were employed. GLS was the standard of reference, while MRL was useful in identifying non-typical values caused by sampling errors or pathological sources. Graphs were used to review the fit, and the software also printed a summary, for each distribution module, of the standard error of estimate (SE) plus pass or fail results in runs and confidence limit tests. In theory, the smallest SE using GLS should have indicated the best fit and it was confirmed by passing both the runs and confidence limit tests. The best fit was also reviewed to assure that it was consistent with the distribution that might be expected from the process. When two or more SEs were quite close, the MRL was used as a secondary screen. A drastic difference in SE between regression models usually suggested the distorting influence of outliers. We have used the data from other years of operation as an indicator for the appropriate distribution (if adjacent years follow the log-normal distribution, a year with sparse data where Weibull and lognormal fit equally well would be selected as log-normal). A detailed listing gave classically calculated mean and standard deviation, estimated parameters for the chosen regression model, regression equation, SE, and results of runs and confidence limit tests.

The Crystal Ball software was a forecasting and risk analysis tool for the Excel spreadsheet software. Crystal Ball was written in Excel Version 4 macro language and extended the standard spreadsheet capabilities. A spreadsheet has two major limitations for risk analysis: only one spreadsheet value (or cell) can be changed at a time, making it difficult to examine a range of outcomes; and the "What-if" solver is a single point estimate which does not indicate the probability of occurrence. Crystal Ball extended the spreadsheet by allowing a range of values,

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described by a distribution, to be placed as the value in a cell. Crystal Ball also permitted Monte Carlo Simulation, in which the distribution was sampled from in a random fashion, displaying the results as a forecast which shows the range of possible outcomes, and their probability, for the range of possibilities associated with the assumptions. This permitted rapid and low cost assembly of a risk assessment model. The monte carlo simulation using Crystal Ball provided a simple way to integrate the distribution and get 95 percentile limits.

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4. ASSUMPTIONS

The following assumptions were made in this calculation:

- no reduction factor was assumed for the ESA pathway due to filters
- no reduction factor was assumed for the DD pathway due to filters or settling

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5. CALCULATION

Data for the ESA pathway for the three buildings K-1131, K-1401, and K-1420 had to be statistically analyzed. The statistical analysis software PDP reported that the extreme value distribution best described the data for K-1131. Table 5.1 reports the parameters used in the Crystal Ball simulation. Fig. 5.1 shows a graph of the distribution. The header in Fig. 5.1 specifies the month of January; all twelve months of the year had this distribution, and only January was taken as an example. The horizontal axis is in units of grams of uranium, and vertical axis gives the normalized probability.

Table 5.1 K-1131 Distribution Parameters

Extreme Value distribution with parameters:

Mode	22374
Scale	16503

Selected range is from 0 to +Infinity

Mean value in simulation was 32692

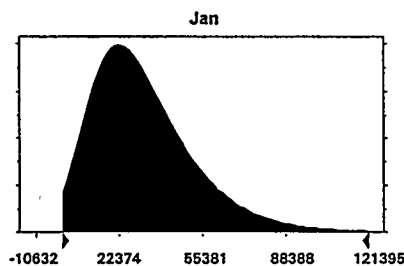


Figure 5.1 K-1131 Release Distribution

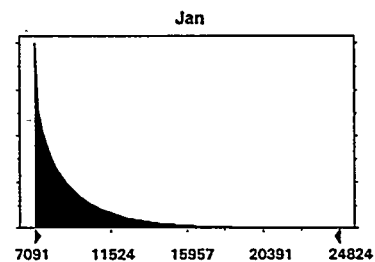


Figure 5.2 K-1401 Release Distribution

In Fig. 5.2 the distribution for the K-1401 is shown. The data were best described by a Weibull distribution, and the parameters for that distribution are given in Table 5.2.

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Table 5.2 K-1401 Distribution Parameters

Weibull distribution with parameters:

Location	7091
Scale	2274
Shape	0.8717

Selected range is from 7091 to +Infinity

Mean value in simulation was 9501

In Fig. 5.3 the distribution for the K-1420 is shown. The data were best described by a log-normal distribution, and the parameters for that distribution are given in Table 5.3.

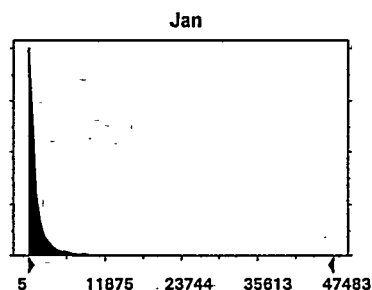


Table 5.3 K-1420 Distribution Parameters

Lognormal distribution with parameters:

Mean	1576
Standard Dev.	4752

Selected range is from 0 to +Infinity

Mean value in simulation was 1587

Figure 5.3 K-1420 Release Distribution

Once twelve cells in an Excel spreadsheet were defined to have the above distributions for each of the buildings, another cell was defined that was the sum of the individual twelve cells, and it represented the yearly release amount. This cell was the forecast cell in the Crystal Ball simulation. During the simulation, the statistical behavior of the yearly release was recorded. Figures 5.4 through 5.6 and Tables 5.4 through 5.6 give the statistical behavior that was recorded. In each case the mean of each distribution became the asserted annual release for the respective building. Although the mode was the most probable value, by definition, for each distribution, using the mean introduced an appropriate amount of conservatism. No reductions due to filter traps or scrubbers were included at this point in the calculation.

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[Signature] 10/31/96
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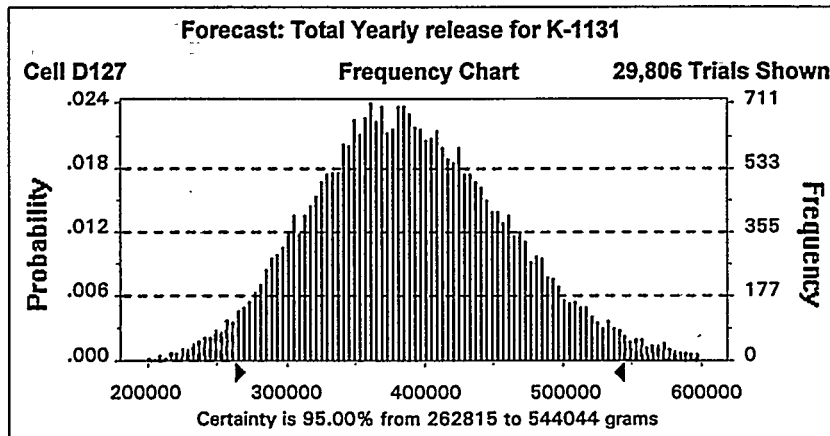


Figure 5.4 K-1131 Annual Release Distribution

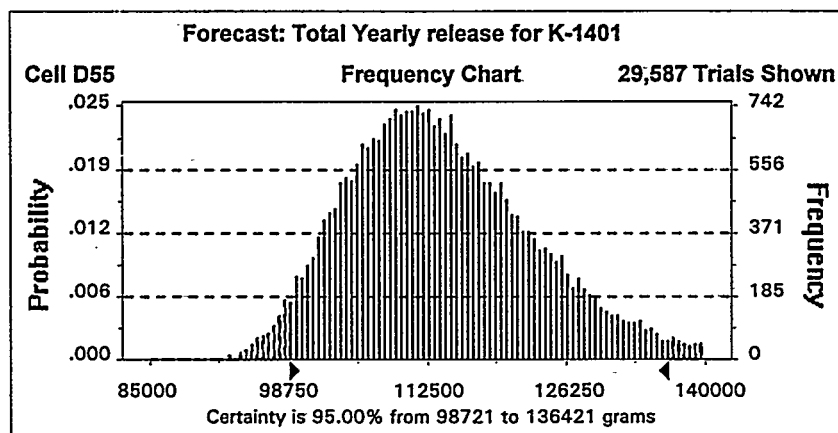


Figure 5.5 K-1401 Annual Release Distribution

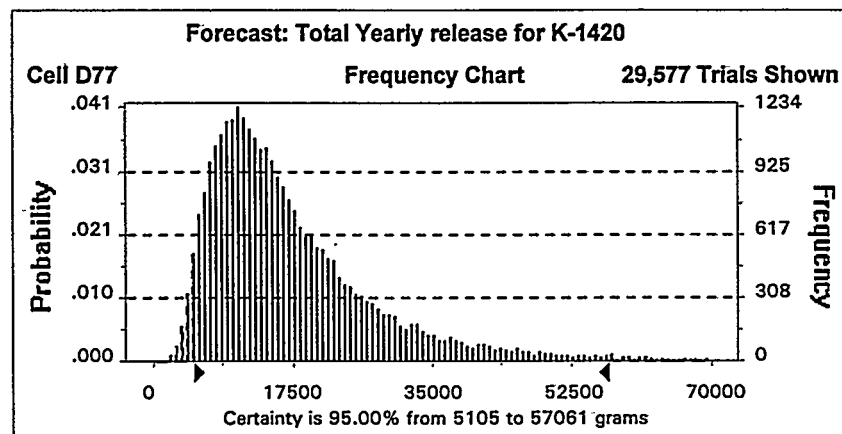


Figure 5.6 K-1420 Annual Release Distribution

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10/31/96Title Fitting Uranium Releases for ESA, EIVA, CIVA, and DD Pathways

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Table 5.4 K-1131 Annual Release Statistics

Forecast: Total Yearly release for K-1131

[MASTER4.XLS]K-1131 - Cell: D127

Summary:

Certainty Level is 95.00%

Certainty Range is from 262815 to 544044 grams

Display Range is from 200000 to 600000 grams

Entire Range is from 145779 to 740785 grams

After 30,000 Trials, the Std. Error of the Mean is 416

Statistics:

	Value
Trials	30000
Mean	391735
Median (approx.)	387248
Mode (approx.)	368906
Standard Deviation	72026
Variance	5187784595
Skewness	0.37
Kurtosis	3.27
Coeff. of Variability	0.18
Range Minimum	145779
Range Maximum	740785
Range Width	595006
Mean Std. Error	415.84

then in two separate calculation
take the 50% and the 95%
we have no data. ~~for~~ For building K-1131
this included 1951 → 1952 (multiplied x 0 - was the plant not operating?)
1953
1954
1955
- all other years that this plant operated
for, we had release data.
1956-1961
~~for K-1140~~

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Table 5.5 K-1401 Annual Release Statistics

Forecast: Total Yearly release for K-1401

[MASTER4.XLS]K-1401 - Cell: D55

Summary:

Certainty Level is 95.00%

Certainty Range is from 98721 to 136421 grams

Display Range is from 85000 to 140000 grams

Entire Range is from 89832 to 167505 grams

After 30,000 Trials, the Std. Error of the Mean is 56

95% Certainty

Statistics:

Trials

Mean

Median (approx.)

Mode (approx.)

Standard Deviation

Variance

Skewness

Kurtosis

Coeff. of Variability

Range Minimum

Range Maximum

Range Width

Mean Std. Error

Value

30000

114332

113183

111969

9662

93354686

0.71

3.75

0.08

89832

167505

77673

55.78

consider this
50%

Then take the ~~95~~ 50th and 95th certainty levels ^{values} and use them for years in which we did not have data. For K-1401 these included the years
[1946-1951 and
1956-1962.

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Table 5.6 K-1420 Annual Release Statistics

Forecast: Total Yearly release for K-1420

[MASTER4.XLS]K-1420 - Cell: D77

Summary:

Certainty Level is 95.00%

Certainty Range is from 5105 to 57061 grams

Display Range is from 0 to 70000 grams

Entire Range is from 2133 to 514484 grams

After 30,000 Trials, the Std. Error of the Mean is 92

Statistics:

Trials

Mean

Median (approx.)

Mode (approx.)

Standard Deviation

Variance

Skewness

Kurtosis

Coeff. of Variability

Range Minimum

Range Maximum

Range Width

Mean Std. Error

Value

30000

18787

14836

9819

15949

254384131

6.40

102.87

0.85

2133

514484

512351

92.08

Then take the 50th and 95th certainty levels and use these values for years that we do not have release data. For K-1420 Now for the cascade purge DD pathway. Figure 5.7 and Table 5.7 give the graph and these years av. distribution parameters for the annual number of releases. Figure 5.8 and Table 5.8 give the graph and distribution parameters for the release mass.

1953-1959

-part of 1963

1964-1985

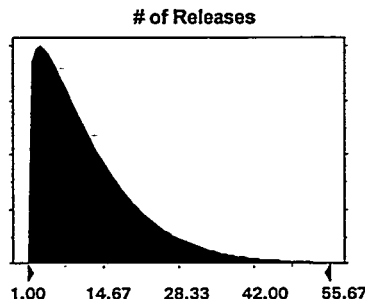


Figure 5.7 Cascade Purge Vaults Annual Number of Releases Distribution

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**Table 5.7 Cascade Purge Vaults' Annual Number of Releases
Distribution Parameters**

Assumption: # of Releases

Weibull distribution with parameters:

Location	1.00
Scale	11.68
Shape	1.16

Selected range is from 1.00 to +Infinity

Mean value in simulation was 12.13

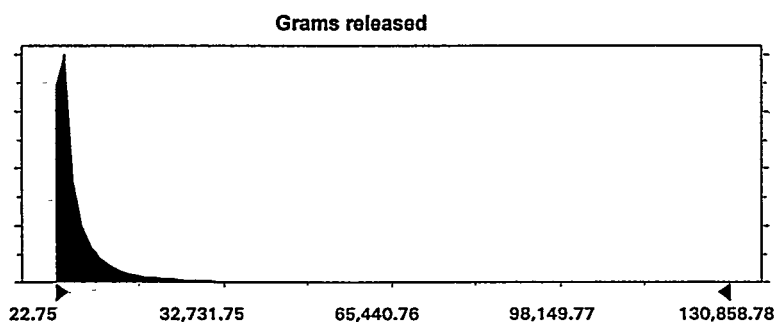


Figure 5.8 Cascade Purge Vaults Grams Released Distribution

**Table 5.8 Cascade Purge Vaults' Grams Released
Distribution Parameters**

Assumption: Grams released

Lognormal distribution with parameters:

Mean	4,886.00
Standard Dev.	12,946.10

Selected range is from 0.00 to +Infinity

Mean value in simulation was 4,799.62

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In an Excel spreadsheet, two cells were defined to have the above properties. Another cell was defined as the product of these two, and it thus represented the total annual release mass for the cascade purge DD pathway. This cell became the forecast cell in a Crystal Ball simulation. The statistical behavior of the forecast is shown in Figure 5.9 and listed in Table 5.9. The resulting distribution was extremely positively skewed as shown in Figure 5.9 and evidenced in Table 5.9. Such a degree of skewness resulted in a mean greater than twice the mode. This is also reflected in the standard deviation given in Table 5.9; the variance is so large that it exceeded the display format and had to be manually entered. To have asserted the mean as the total annual release mass would have been excessively conservative, and thus the mode was asserted as the annual release mass.

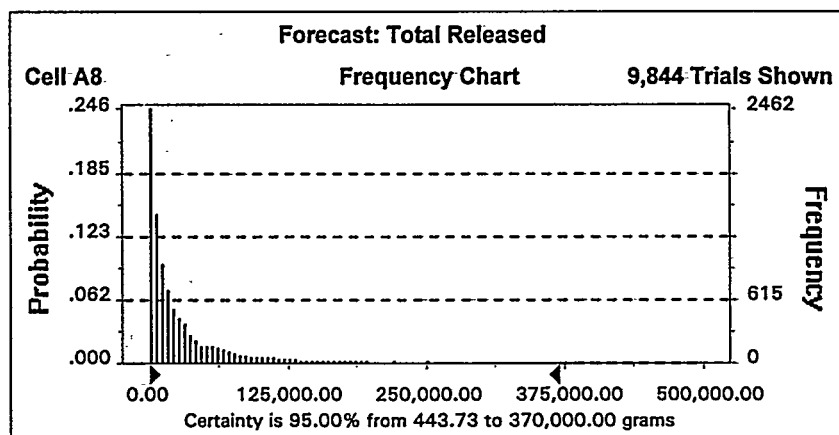


Figure 5.9 Annual Release Distribution for Cascade Purge Vaults by DD Pathway

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Table 5.9 Annual Release Distribution Parameters for Cascade Purge Vaults**Forecast: Total Released****Summary:**

Certainty Level is 95.00%

Certainty Range is from 443.73 to 370,000.00 grams

Display Range is from 0.00 to 500,000.00 grams

Entire Range is from 8.05 to 5,171,283.16 grams

After 10,000 Trials, the Std. Error of the Mean is 1,759.90

Statistics:

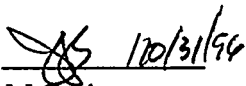
	<u>Value</u>
Trials	10000
Mean	57,243.28
Median (approx.)	16,601.80
Mode (approx.)	25,864.43
Standard Deviation	175,989.81
Variance	30,972,411,564.55
Skewness	13.11
Kurtosis	268.40
Coeff. of Variability	3.07
Range Minimum	8.05
Range Maximum	5,171,283.16
Range Width	5,171,275.11
Mean Std. Error	1,759.90

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6. REFERENCES

SRA-95-002	Bennett, T.E. Uranium Release Estimates for the ORGDP Purge Cascade 1953-1955. 1995.
SRA-95-011	Bennett, T.E. Uranium Release Estimates for the ORGDP Purge Cascade 1961. 1995.
SRA-95-012	Bennett, T.E. Uranium Release Estimates for the ORGDP Purge Cascade 7/68 - 6/69. 1995.
SRA-95-013	Bennett, T.E. Uranium Release Estimates for the ORGDP Purge Cascade 7/75 - 6/76. 1995.
SRA-96-009	Burmeister, R.E. Fitting Uranium Release Estimates of the Purge Cascade. 1996.
SRA-96-012	Burmeister, R.E. The Master Release List and Source Term for K-25
TEAM	Probability Distribution Plotting (PDP) Software Version 3.21 for DOS; available from Technical and Engineering Aids for Management (TEAM), Box 25, Tamworth, NH 03866
Crystal	Crystal Ball Version 3.0 from Decisioneering, Inc. 1380 Lawrence Street, Suite 520, Denver, CO 80204-9849

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7. ELECTRONIC FILES

The following files are included on the diskette that accompanies this calculation.

<u>File Name</u>	<u>Description</u>
SRA010.DOC	This calculation in MS-WORD format
CIVA.XLS	EXCEL spreadsheet for CIVA pathway work
DD.XLS	EXCEL spreadsheet for DD pathway work
DDRPT.XLS	EXCEL spreadsheet for Crystal Ball work for DD pathway
EIVA.XLS	EXCEL spreadsheet for EIVA pathway work
MASTER4.XLS	EXCEL spreadsheet for ESA pathway work
REPORT2.XLS	EXCEL spreadsheet for Crystal Ball work for ESA pathway

All of the following files are the output from the PDP software. Nomenclature is as follows: DDALL.* are the PDP output files for the DD pathway work; K_ESA1.* are the output files for the K-1131 building for the ESA pathway; K_ESA2.* are the output files for the K-1401 building for the ESA pathway, and K_ESA3.* are the output files for the K-1420 building for the ESA pathway. The *.RAW are the data files used by PDP which were taken from the above spreadsheets.

DDALL.CMP
DDALL.HIS
DDALL.L
DDALL.LGE
DDALL.LGN
DDALL.P
DDALL.R
DDALL.RAW
DDALL.RC
DDALL.S\$
DDALL.S1
DDALL.S2
DDALL.S3
DDALL.S4
DDALL.S5
DDALL.SC
DDALL.WEI
K_ESA1.CMP
K_ESA1.HIS
K_ESA1.L

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K_ESA1.P
K_ESA1.R
K_ESA1.RAW
K_ESA1.RC
K_ESA1.RP1
K_ESA1.S\$
K_ESA1.S1
K_ESA1.S2
K_ESA1.S3
K_ESA1.S4
K_ESA1.S5
K_ESA1.SC
K_ESA2.CMP
K_ESA2.HIS
K_ESA2.K_E
K_ESA2.L
K_ESA2.P
K_ESA2.R
K_ESA2.RAW
K_ESA2.RC
K_ESA2.RP1
K_ESA2.RP2
K_ESA2.S\$
K_ESA2.S1
K_ESA2.S2
K_ESA2.S3
K_ESA2.S4
K_ESA2.S5
K_ESA2.SC
K_ESA3.CMP
K_ESA3.HIS
K_ESA3.L
K_ESA3.P
K_ESA3.R
K_ESA3.RAW
K_ESA3.RC
K_ESA3.RP1
K_ESA3.S\$
K_ESA3.S1
K_ESA3.S2
K_ESA3.S3

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K_ESA3.S4

K_ESA3.S5

K_ESA3.SC

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CALC NO SRA-96-012 REV 1

Project/Task ChemRisk/TDH Oak Ridge Dose Reconstruction

Prepared by R. E. Burmeister Date 2/11/97 Checked by/Date J. J. Shonka

Title The Atmospheric Master Release List and Atmospheric Source Term for K-25

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#3189

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Calculation Control Sheet

Calculation number: SRA-96-012 REV. 1

Title: The Atmospheric Master Release List and Atmospheric Source Term for K-25

Reason for calculation/revision: correction of error in calculation, pages 5, 7, 8, and 17 affected

Client: ChemRisk/TDH

Project: Oak Ridge Dose Reconstruction

Project/Task Number: Task 6

Prepared by: Regan E. Burmeister
Regan E. Burmeister (signed original on file)

Date: 2/11/97

Independent Technical Review by: Joseph J. Shonka

Joseph J. Shonka (signed original on file)

Date: 2/13/97

Quality Assurance Review by: Deborah B. Shonka

Deborah B. Shonka (signed original on file)

Date: 2/13/97



This calculation has been voided or superseded by

(calculation number)

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Review Method Sheet

The undersigned has reviewed this calculation in accordance with the method(s) indicated below.

1. Computer Aided Calculation	
a	Review to determine that the computer program(s) has been validated and documented, is suitable to the problem being analyzed, and that the calculation contains all necessary information for reconstruction at a later date.
b	Review to determine that the input data as specified for program execution is consistent with the design input, correctly defines the problem for the computer algorithm and is sufficiently accurate to produce results within any numerical limitations of the program.
c	Review to verify that the results obtained from the program are correct and within stated assumptions and limitations of the program and are consistent with the input.
d	Review validation documentation for temporary changes to listed, or developmental, or unique single application programs, to assure that the methods used adequately validate the program for the intended application.
e	Review of code input only, since the computer program has sufficient history of use at Shonka Research Associates, Inc. in similar calculations.
f	Review arithmetic necessary to prepare code input data.
g	Other:
2. Hand Prepared Calculations	
a	Detailed review of the original calculations.
b	Review by an alternate, simplified, or approximate method of calculation.
c	Review of a representative sample of repetitive calculations.
d	Review of the calculation against a similar calculation previously performed.
e	Other:
3. Revisions	
a	Editorial changes only
b	Elimination of unapproved input data without altering calculated results.
©	Other: updated input data to present version of release values which altered calculated results
4. Other	

Reviewer: _____

Joseph J. Shonka (signed original on file)

Date: _____

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CALC NO SRA-96-012 REV 1

Project/Task ChemRisk/TDH Oak Ridge Dose Reconstruction

Prepared by R. E. BurmeisterDate 2/11/97

Checked by/Date

J. J. Shonka

Title The Atmospheric Master Release List and Atmospheric Source Term for K-25

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ABSTRACT

This calculation presents the ORHS-II Atmospheric Master Release List for the Oak Ridge Gaseous Diffusion Plant. The list is an Excel spreadsheet with many columns and sheets that detail the construction of the uranium release history from 1945 to 1995. All of the data used in the list are given references. The formulas for certain columns are explained. In particular, the use of the results of previous calculations to fill chronological gaps in the release history are detailed.

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Title The Atmospheric Master Release List and Atmospheric Source Term for K-25

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3. METHODS	9
4. ASSUMPTIONS	10
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Appendix A Copies of two personal communications
Task 6: Review of Release Fraction Literature
1986-1995 K-25 Uranium Airborne Releases

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1. INTRODUCTION

Task 6 of the Oak Ridge Dose Reconstruction focuses on the evaluation of the quality of historical airborne and waterborne effluent monitoring data and the determination of the potential significance of unmonitored emissions. Uranium played an important role throughout historical operations on the Oak Ridge Reservation (ORR) and is known to have been released to the environment through air and water. The two largest uses of uranium on the Reservation were the enrichment processes of the ^{235}U isotope by electromagnetic separation at the Y-12 facility and gaseous diffusion at the K-25 facility.

This calculation focused on atmospheric uranium releases from the gaseous diffusion process at the K-25 site. The K-25 site was comprised of the five different cascade complexes, K-25, K-27, K-29, K-31, and K-33, as well as many buildings that supported the gaseous diffusion process. Included as part of the K-25 site for this calculation was the S-50 liquid thermal diffusion plant (SRA-96-011). Many releases were accidents due to equipment failures or personnel mistakes. Other releases were scheduled and deliberate, such as the releases from the purge cascade. Releases through other pathways that did not include the atmosphere were collected and documented during the search for material release events. These releases were not included in this calculation.

As much information as possible about each release was gathered. Releases were typified by their date of occurrence and amount of release as well as other information that allowed the releases to be classed according to release pathway or location of release. It was known that certain gaps in the historical releases from certain buildings existed; release data was unavailable or did not exist but these buildings were operational for known time frames. The results of two previous calculations, SRA-96-009 and SRA-96-010, were used to fill these gaps.

Revision 1 of this calculation differs from revision 0 in that an examination of the master spreadsheets ATM10A.XLS and ATM10B.XLS revealed that data used in the calculation of environmental releases (see sheet *Environment*) and in the calculation of differences (see sheet *10% Diff*) was not current to the present version of the release values. These errors were corrected, and the spreadsheets were updated to versions ATM11A.XLS and ATM11B.XLS. The corrections resulted in an approximate 24 kg and 12 kg increase in the total uranium release at the 50%-ile and 95%-ile.

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2. SUMMARY OF RESULTS

Table 2.1 and Table 2.2 give the chronological atmospheric uranium release history for the Oak Ridge Gaseous Diffusion Plant (ORGDP) for the 50% and 95% certainty values, respectively. Care should be taken in using these values.

The two certainty values were used to bound the release values. These certainty values were the results of statistical fits to some of the release data (SRA-96-009 and SRA-96-010). In all other cases, when releases were not fit, no uncertainty was asserted. Thus, the actual uncertainty in the release results was larger than has been expressed in this calculation. Only releases whose pathway was evaluated as being released to the atmosphere were included here.

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Table 2.1 Uranium Release History for 50% Certainty

Atmospheric Pathways Yearly Release									
Date	Uranium (kg)	Uranium (Ci)	U-235 (kg)	U-238 (kg)	U-235 (Ci)	U-238 (Ci)	U-234 (Ci)	Uranium Cumulative (kg)	Uranium Cumulative (Ci)
1944	412.5	0.27716	2.9	409.6	0.00634	0.13766	0.13316	412.5	0.27716
1945	1246.7	0.83816	8.9	1237.8	0.01918	0.41605	0.40292	1659.2	1.11532
1946	115.8	0.19671	3.4	112.4	0.00729	0.03778	0.15163	1775.0	1.31202
1947	115.5	0.21226	3.5	112.0	0.00754	0.03764	0.16708	1890.5	1.52428
1948	119.9	0.20789	3.4	116.4	0.00732	0.03912	0.16145	2010.4	1.73217
1949	192.9	0.24933	3.7	189.1	0.00802	0.06356	0.17774	2203.2	1.98150
1950	250.8	0.29517	4.3	246.1	0.00930	0.08272	0.20315	2454.0	2.27667
1951	707.4	0.45399	7.3	700.0	0.01587	0.23528	0.20284	3161.4	2.73066
1952	1211.7	0.83800	8.6	1203.1	0.01854	0.40438	0.41508	4373.1	3.56866
1953	1307.0	1.20957	17.2	1289.8	0.03726	0.43351	0.73880	5680.1	4.77823
1954	459.1	0.98574	15.4	443.7	0.03325	0.14912	0.80337	6139.1	5.76396
1955	482.7	0.40827	6.1	476.6	0.01313	0.16020	0.23493	6621.9	6.17223
1956	397.1	0.45520	6.5	390.7	0.01396	0.13132	0.30993	7019.0	6.62743
1957	442.8	0.50339	7.1	435.7	0.01540	0.14645	0.34154	7461.8	7.13082
1958	2711.0	2.11029	25.5	2685.0	0.05515	0.90248	1.15266	10172.8	9.24111
1959	675.8	0.88153	12.7	661.9	0.02735	0.22248	0.63171	10848.6	10.12264
1960	1189.6	0.89979	10.7	1178.9	0.02307	0.39624	0.48048	12038.1	11.02243
1961	896.9	0.75221	8.9	888.0	0.01932	0.29846	0.43443	12935.1	11.77464
1962	163.5	0.34328	6.0	157.5	0.01288	0.05295	0.27744	13098.6	12.11792
1963	1005.0	5.15180	113.1	891.9	0.24446	0.29977	4.60757	14103.6	17.26972
1964	23.6	0.09661	1.8	21.8	0.00379	0.00733	0.08549	14127.2	17.36634
1965	456.6	0.90349	17.6	439.0	0.03798	0.14756	0.71794	14583.8	18.26983
1966	19.4	0.09083	1.7	17.7	0.00361	0.00595	0.08128	14603.1	18.36066
1967	18.8	0.09004	1.7	17.1	0.00358	0.00576	0.08070	14622.0	18.45071
1968	20.6	0.09107	1.7	18.9	0.00360	0.00637	0.08111	14642.6	18.54178
1969	28.8	0.09718	1.7	27.1	0.00376	0.00910	0.08432	14671.4	18.63896
1970	24.8	0.09672	1.8	23.0	0.00382	0.00774	0.08516	14696.2	18.73567
1971	70.1	0.18272	3.5	66.4	0.00753	0.02232	0.15287	14766.2	18.91839
1972	49.0	0.12644	2.3	46.7	0.00492	0.01570	0.10582	14815.2	19.04483
1973	284.5	0.44000	9.5	275.0	0.02046	0.09243	0.32711	15099.7	19.48483
1974	622.0	1.52140	33.0	589.0	0.07124	0.19796	1.25220	15721.7	21.00623
1975	371.0	0.78680	17.2	353.8	0.03713	0.11891	0.63076	16092.7	21.79303
1976	114.4	0.25000	5.9	110.7	0.01275	0.03722	0.20003	16207.1	22.04303
1977	36.5	0.12694	2.4	34.0	0.00521	0.01144	0.11029	16243.6	22.16997
1978	28.1	0.10481	1.9	26.1	0.00418	0.00879	0.09183	16271.7	22.27478
1979	46.1	0.11000	3.0	43.1	0.00653	0.01448	0.08899	16317.8	22.38478
1980	121.7	0.20000	4.8	116.9	0.01037	0.03929	0.15034	16439.5	22.58478
1981	68.7	0.13000	3.5	65.1	0.00762	0.02189	0.10049	16508.2	22.71478
1982	73.8	0.11000	3.2	70.6	0.00691	0.02373	0.07937	16582.0	22.82478
1983	19.5	0.09116	1.7	17.8	0.00363	0.00598	0.08155	16601.4	22.91594
1984	19.3	0.09068	1.7	17.5	0.00361	0.00588	0.08119	16620.7	23.00662
1985	20.0	0.09059	1.7	17.4	0.00360	0.00586	0.08113	16640.6	23.09721
1986	0.2	0.00100	0.0	0.2	0.00005	0.00006	0.00089	16640.8	23.09821
1987	0.4	0.00030	0.0	0.4	0.00001	0.00013	0.00016	16641.2	23.09851
1988	1.7	0.00110	0.0	1.7	0.00002	0.00057	0.00051	16642.9	23.09961
1989	1.1	0.00040	0.0	1.1	0.00000	0.00037	0.00003	16644.1	23.10001
1990	2.0	0.00110	0.0	2.0	0.00002	0.00067	0.00041	16646.1	23.10111
1991	40.2	0.02400	0.2	40.0	0.00045	0.01345	0.01010	16686.3	23.12511
1992	112.4	0.06400	0.5	111.9	0.00108	0.03761	0.02531	16798.7	23.18911
1993	12.0	0.01000	0.1	11.9	0.00029	0.00399	0.00572	16810.7	23.19911
1994	10.0	0.00800	0.1	9.9	0.00023	0.00333	0.00445	16820.7	23.20711
1995	16.2	0.00670	0.0	16.2	0.00001	0.00545	0.00123	16836.9	23.21381
Totals	16836.9	23.21381	399.2	16436.4	0.86261	5.52451	16.82670		

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Table 2.2 Uranium Release History at 95% Certainty

Atmospheric Pathways Yearly Release									
Date	Uranium (kg)	Uranium (Ci)	U-235 (kg)	U-238 (kg)	U-235 (Ci)	U-238 (Ci)	U-234 (Ci)	Uranium Cumulative (kg)	Uranium Cumulative (Ci)
1944	1287.5	0.86507	9.2	1278.3	0.01978	0.42967	0.41562	1287.5	0.86507
1945	3871.7	2.60189	27.5	3844.2	0.05952	1.29208	1.25029	5159.2	3.46696
1946	137.9	0.37671	3.9	134.0	0.00836	0.04504	0.32330	5297.1	3.84367
1947	138.4	0.44751	4.8	133.6	0.01032	0.04491	0.39228	5435.5	4.29118
1948	142.9	0.44313	4.7	138.0	0.01009	0.04639	0.38665	5578.3	4.73431
1949	215.8	0.48457	5.0	210.8	0.01080	0.07084	0.40294	5794.1	5.21889
1950	273.7	0.53041	5.6	267.7	0.01208	0.08999	0.42835	6067.8	5.74930
1951	882.6	0.72924	9.7	872.9	0.02092	0.29339	0.41493	6950.4	6.47854
1952	1212.5	0.89325	9.4	1203.2	0.02024	0.40440	0.46861	8162.9	7.37179
1953	1287.2	1.48746	18.9	1268.3	0.04076	0.42630	1.02041	9450.1	8.85925
1954	637.0	1.45242	19.7	617.3	0.04262	0.20748	1.20233	10087.1	10.31167
1955	597.2	0.85827	10.0	587.2	0.02155	0.19735	0.63936	10684.3	11.16993
1956	458.3	1.12045	11.1	447.2	0.02402	0.15032	0.94611	11142.6	12.29038
1957	504.0	1.16864	11.8	492.3	0.02546	0.16545	0.97772	11646.7	13.45902
1958	2711.0	2.80819	31.6	2678.9	0.06839	0.90042	1.83938	14357.7	16.26721
1959	737.0	1.54678	17.3	718.5	0.03741	0.24148	1.26789	15094.6	17.81399
1960	1211.8	1.08781	11.3	1200.5	0.02439	0.40350	0.65991	16306.4	18.90179
1961	919.0	0.93221	9.4	909.6	0.02039	0.30572	0.60610	17225.4	19.83400
1962	185.8	0.53130	6.6	179.1	0.01420	0.06021	0.45688	17411.1	20.36530
1963	1005.0	4.99809	105.1	899.9	0.22712	0.30247	4.46850	18416.1	25.36339
1964	61.9	0.52662	5.1	56.7	0.01108	0.01907	0.49647	18478.0	25.89001
1965	456.6	1.32942	21.1	435.6	0.04550	0.14639	1.13752	18934.6	27.21943
1966	57.6	0.52084	5.0	52.6	0.01090	0.01768	0.49226	18992.2	27.74028
1967	57.1	0.52005	5.0	52.1	0.01087	0.01750	0.49169	19049.3	28.26033
1968	58.9	0.52108	5.0	53.9	0.01088	0.01810	0.49209	19108.2	28.78140
1969	67.1	0.52718	5.1	62.0	0.01104	0.02083	0.49531	19175.3	29.30858
1970	63.1	0.52672	5.1	57.9	0.01111	0.01947	0.49615	19238.4	29.83531
1971	108.4	0.61272	6.9	101.3	0.01481	0.03405	0.56385	19346.7	30.44803
1972	82.3	0.54491	5.4	76.9	0.01169	0.02585	0.50737	19429.0	30.99293
1973	284.5	0.57137	11.7	272.8	0.02532	0.09168	0.45438	19713.5	31.56431
1974	622.0	2.66417	52.5	569.4	0.11348	0.19139	2.35930	20335.5	34.22848
1975	371.0	1.45877	26.2	344.7	0.05672	0.11587	1.28619	20706.5	35.68725
1976	114.4	0.61003	7.5	109.1	0.01630	0.03667	0.55706	20820.9	36.29727
1977	74.7	0.55694	5.8	68.9	0.01249	0.02317	0.52127	20895.7	36.85421
1978	66.4	0.53481	5.3	61.1	0.01147	0.02052	0.50282	20962.0	37.38902
1979	59.8	0.52658	5.2	54.7	0.01116	0.01837	0.49705	21021.9	37.91560
1980	121.7	0.52851	7.0	114.7	0.01503	0.03856	0.47491	21143.5	38.44410
1981	68.7	0.52667	5.4	63.2	0.01177	0.02125	0.49366	21212.2	38.97077
1982	73.8	0.52006	5.5	68.3	0.01188	0.02295	0.48523	21286.0	39.49084
1983	57.7	0.52117	5.0	52.7	0.01091	0.01771	0.49254	21343.8	40.01201
1984	57.5	0.52069	5.0	52.4	0.01089	0.01761	0.49218	21401.3	40.53270
1985	58.2	0.52060	5.0	52.3	0.01089	0.01759	0.49212	21459.5	41.05329
1986	0.2	0.00100	0.0	0.2	0.00005	0.00006	0.00089	21459.7	41.05429
1987	0.4	0.00030	0.0	0.4	0.00001	0.00013	0.00016	21460.1	41.05459
1988	1.7	0.00110	0.0	1.7	0.00002	0.00057	0.00051	21461.8	41.05569
1989	1.1	0.00040	0.0	1.1	0.00000	0.00037	0.00003	21462.9	41.05609
1990	2.0	0.00110	0.0	2.0	0.00002	0.00067	0.00041	21464.9	41.05719
1991	40.2	0.02400	0.2	40.0	0.00045	0.01345	0.01010	21505.2	41.08119
1992	112.4	0.06400	0.5	111.9	0.00108	0.03761	0.02531	21617.6	41.14519
1993	12.0	0.01000	0.1	11.9	0.00029	0.00399	0.00572	21629.6	41.15519
1994	10.0	0.00800	0.1	9.9	0.00023	0.00333	0.00445	21639.6	41.16319
1995	16.2	0.00670	0.0	16.2	0.00001	0.00545	0.00123	21655.8	41.16989
Totals	21655.8	41.16989	544.5	21109.9	1.17676	7.09534	32.89779		

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Project/Task ChemRisk/TDH Oak Ridge Dose Reconstruction

Prepared by R. E. Burmeister

Date 2/11/97

Checked by/Date J. J. Shonka

Title The Atmospheric Master Release List and Atmospheric Source Term for K-25

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3. METHODS

An Excel spreadsheet was developed that primarily functioned as a database of uranium releases from the ORGDP. Documents were retrieved from record centers at the Oak Ridge Reservation (ORR). The documents were mainly accountability records that indicated when, where, why, what and how much material containing uranium was released. This information was entered into the database.

Interviews conducted with active and retired personnel help to confirm the type and scope of activities that occurred during their employment (Bennett 1995A)(Bennett 1995B)(Bennett 1995C)(Buddenbaum 1995)(Burmeister 1996)(Shonka 1995). Using this information and other historical documents, it could be determined whether or not the database had a complete release history for a particular ORGDP building. For those high priority buildings that did not have a complete history, reasonable estimates of releases were made using probability distributions (SRA-96-009, -010, -011). These estimates were added to the database for the particular time period during which they were applicable.

An annual release amount in kilograms and curies of uranium was determined by summing all release amounts for each year of operation from 1944 to 1995. These are the release amounts given in Tables 2.1 and 2.2.

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4. ASSUMPTIONS

The following assumptions were made in this calculation:

- 1) The Environmental Monitoring Reports for the Oak Ridge facilities reported curies released to the atmosphere. It was assumed that these releases were due entirely to the ORGDP (See *Environment* in Section 5.) This has overstated the releases in the post 1980 time frame.
- 2) It was assumed that the building release fraction for all buildings was unity; i.e., all releases inside buildings that had a pathway to the atmosphere were assumed to transport 100% to the atmosphere. (Appendix A, JJS.048*).
- 3) No corrections were made for sample line loss. Losses estimated from stack sampling may be significantly understated, perhaps by as much as a factor of 4 or more.
- 4) The trapping efficiency for the purge cascade releases was assumed to 85%; i.e. 15% of material was released to the atmosphere.(SRA-96-009).
- 5) Natural enrichment of 0.711% was assumed for the environmental release data of 1989. Release data were inconsistent to provide a proper yearly enrichment level.

* Private communication titled, "Task 6: Review of Release Fraction Literature". Copies to Tom Widner and Jack Buddenbaum. 8/22/96.

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5. CALCULATION

This section details how the Atmospheric Master Release List (the List) spreadsheets were constructed. The reader should refer to the electronic copy on the enclosed disk. The List has twelve worksheets titled as one scrolls left to right as follows: 1) *Atmospheric Release*, 2) *Atm. Yearly Release*, 3) *Cylinder Fire Test*, 4) *Cascade Fit (ESA)*, 5) *K-1131, K-1401, K-1420 Fits (ESA)*, 6) *Environment*, 7) *Environment 2* 8) *10% Diff*, 9) *S-50*, 10) *Uranium*, 11) *New Data 10-31-96*, and 12) *New Data 8-29-96*. The descriptions of these sheets given below apply to both the 50% and 95% certainty fit values that were determined in the previous calculations. There are actually two spreadsheets; one for 50% certainty and one for 95% certainty. The release data are the same in the two spreadsheets; only the fit values change between the spreadsheets.

The worksheet *Atmospheric Release* contains a chronological listing of releases of uranium for the ORGDP. Releases are classified on their location, amounts of uranium, U-238, and U-235, weight percents U-235, and release pathways. Some descriptive notes and references are given for each release. This worksheet was assembled primarily from accountability records that were retrieved from records centers on the ORR. Releases were assessed into several pathways. The ESA pathway was used to describe releases from Equipment to Stacks or vents and thus to the Atmosphere. Other pathway categories were described in SRA-96-010.

The worksheet *Atm. Yearly Release* gives the total yearly release amounts for 1945 to 1995 in grams and curies of uranium, U-238, and U-235; curies of U-234 are also given. Cumulative totals are also given along with plots of the release amounts. Entries for a particular year are the sums of many terms. The releases listed in *Atmospheric Release* were summed for each individual year. To these sums were added contributions from *Cascade Fit (ESA)*, *K-1131, K-1401, K-1420 Fits (ESA)*, *Environment*, *Environment 2*, *10% Diff*, *S-50*, *New Data 10-31-96*, and *New Data 8-29-96*. These contributions were estimates of releases that were used to fill in gaps in the release history of the ORGDP for those years that data was unavailable or did not exist. In particular, the releases in *Cascade Fit (ESA)* and *K-1131, K-1401, K-1420 Fits (ESA)*, derived in SRA-96-009 and SRA-96-010, fill in the release history for the purge cascade and the buildings K-1131, K-1401, and K-1420. *New Data 10-31-96* contains releases that were received and reviewed at Shonka Research Associates (SRA) on October 31, 1996. *New Data 8-29-96* contains releases that were received and reviewed at Shonka Research Associates (SRA) on August 29, 1996. This data arrived after much of the work in assembling the List had already been completed.

Cascade Fit (ESA) gives the fitted release values for the purge cascade for all years of operation. Some years have null values; these are the years for which purge cascade data existed. A null value implies that nothing more needed to be added to the yearly release amounts. The assumption of a 15% release fraction, per Assumption 4), was applied here.

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K-1131, K-1401, K-1420 Fits (ESA) gives the fits to the K-1131, K-1401, and K-1420 buildings for the ESA pathway. The fitted values are shown along with factors that represent fractions of years. Some of the release data for these buildings had only a few months of releases for particular years. To fill in the rest of the year, the fractions multiplied the fit values. The results were added to the yearly release amounts. No information was found that indicated that the buildings' operations were any different or were shut down for those missing fractions of years. The release data were only unavailable or did not exist.

The next sheet, *Cylinder Fire Test*, records the releases of UF_6 that occurred in October 1965 as a part of UF_6 cylinder test and development. These releases occurred at the ORGDP Rifle Range and were regarded separately from gaseous diffusion process releases. These releases were added to yearly totals only after releases from *Environment* and *10% Diff*.

The sheet *Environment* gives the atmospheric discharges in curies of uranium for the years 1973 to 1982. The discharges were taken from Environmental Monitoring Reports for the Oak Ridge Facilities for the years 1973 to 1982 inclusive (US AEC 1973)(US ERDA 1974-1976)(US DOE 1977-1982). These reports provided all uranium released from all Oak Ridge Facilities. It was conservatively assumed that all releases were from K-25 operations since there was no way to separate the contributions from all facilities. The curies of uranium were converted into curies and grams of U-238 and U-235. These amounts were then compared to the amounts that had been determined up to this point. Where the difference between the environmental amount and the amount to this point was positive for a particular year, it was assumed that information was missing from the release history and the difference was added to that year's amount. This practice overstated the releases from K-25. The additions occurred on the sheet *Atm. Yearly Release*.

In *Environment* it was necessary to have an enrichment level in order to convert curies to grams. The environmental reports did not give any information regarding enrichment levels. For those years that had a positive difference, the release data was examined to determine the enrichment level. Two methods were used to get an enrichment level indicative of those years. In the first method, the release data for those years were examined to get an average enrichment level; in each year, each data point's enrichment level was summed to a total and then divided by the number of data points. In the second method, each data point's U-235 mass and uranium mass were summed to totals, and then the total U-235 mass was divided by the total uranium mass to get an enrichment level indicative of each year. These two methods are identified in *Environment*. The actual calculation of these enrichments occurred in the spreadsheet ENVIRO.XLS to which *Environment* contained a data link to these enrichments. The second method was chosen because the ratio of masses was the correct definition of a year's enrichment level. In this manner, yearly release data was reconciled with the environmental reports as far as Assumption 1) was concerned (all ORR site releases were due to K-25).

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The sheet *Environment 2* gives environmental data from 1986 to 1995. The sheet contains the original environmental release data for K-25. The release data were kilograms and curies of uranium for the ten years 1986 through 1995. To get a breakdown of kilograms and curies for the nuclides U-238, U-235, and U-234, it was necessary to have the enrichment level. This was calculated by using an expression of alpha specific activity as a function of enrichment (EGG-2530). The resulting enrichment level was an expression of the yearly enrichment, but it was recognized that the values were not average enrichments since no data was available to assert average enrichments. One particular year, 1989, had a calculated enrichment that was negative. This implied that the release data for that year were inconsistent. Since only a small amount of uranium was reported that year, namely 1.11 kg, a natural enrichment of 0.711% was assumed. For such a small release amount, the assumption had a negligible effect on the site cumulative release, but the assumption was the determining factor for that year since the environmental release was the only datum for the year of 1989.

There was another inconsistency in the data for 1992. One reference* reported the release for 1992 as 112.39 kg of uranium at 0.0640 Curies of activity (Appendix A - Buddenbaum Memo). The environmental release report for 1995 reported five years of data and gave the 1992 release amounts as 14.49 kg of uranium at 0.0100 Curies of activity (ENVN-95). The larger values were used in this calculation in order to conservatively state the release for 1992.

The sheet *New Data 10-31-96* contains release data received and reviewed at SRA after primary work had already been completed in assembling yearly releases. No impact of this data was found on any of the prior work. This was mainly due to the releases having release amounts of only a few tens of grams of uranium. The data were added to the yearly releases.

The sheet *New Data 8-29-96* contains release data received at SRA after primary work had already been completed in assembling the yearly releases. This data was reviewed to see if it impacted any of the fitting work accomplished for the purge cascade, K-1131, K-1420, or K-1401. It was determined that there was no impact, and that the data could be added to yearly releases without modifying the previous work.

The yearly release totals to this point were next compared to the reported yearly release amounts from K/HS-95. For those years where the reported value was 10% or more greater than the determined value, the difference was added to the determined value. It was argued that for those years, K/HS-95 had valid but unavailable data that had not yet been retrieved, and to account for such valid data, the differences were added. This analysis occurred on the sheet *10% Diff*.

* Private communication titled, "1986-1995 K-25 Uranium Airborne Releases," from Jack Buddenbaum to Joseph Shonka with copies to Tom Widner and Jennifer Cockroft. 10/23/96.

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The sheet *S-50* contains the releases estimates for the S-50 liquid thermal diffusion plant (SRA-96-011). S-50 operated from September 1944 to September 1945. It produced only low enriched uranium, never exceeding 1% enrichment in product.

The sheet *Uranium* gives physical data for the element uranium and its isotopes. This data was used as needed to calculate grams and curies.

Once all the above had been accomplished, the fit values were replaced with their 95% certainty values determined in the previous calculations. This resulted in two master spreadsheets called ATM11A.XLS and ATM11B.XLS which are included in the disks. ATM11A.XLS contains the 50% certainty work; ATM11B.XLS contains the 95% certainty work.

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6. REFERENCES

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Bennett 1995B	Interview Notes: Jim Rogers. 10/24/95. Inmagic # 2532
Bennett 1995C	Interview Notes: Joe Dykstra. 10/12/95. Inmagic # 2858
Buddenbaum 1995	Interview Notes: Dave Stoddard. 10/11/95. Inmagic # 2533
Burmeister 1996	Interview Notes: Jim Rogers. 9/10/96. Inmagic # 2927
EGG-2530	Department of Energy. Health Physics Manual of Good Practices for Uranium Facilities. EGG-2530. Idaho National Engineering Laboratory. 1988. p. 2-7
ENVN-95	Oak Ridge Reservation Annual Site Environmental Report for 1995. Chapter 3. Table 4.3, Fig. 4.7 and Fig. 4.8. Inmagic #2977
K/HS-95	Lay, A.C. and Rogers, J.G. Oak Ridge Gaseous Diffusion Plant Historical Uranium and Radionuclide Release Report. ORGDP. Martin Marietta. Inmagic #1334
Shonka 1995	Interview Notes: Tom Hanrahan. 10/95. Inmagic # 2534
SRA-96-009	Burmeister, R.E. Fitting Uranium Release Estimates of the Purge Cascade. 1996.
SRA-96-010	Burmeister, R.E. Fitting Estimates for ESA, EIVA, CIVA, and DD Pathways. 1996.
SRA-96-011	Release Estimates for the S-50 Liquid Thermal Diffusion Plant. 1996.

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Project/Task ChemRisk/TDH Oak Ridge Dose Reconstruction

Prepared by R. E. Burmeister

Date 2/11/97

Checked by/Date J. J. Shonka

Title The Atmospheric Master Release List and Atmospheric Source Term for K-25

(signed original on file)

7. ELECTRONIC FILES

The following files are included on the diskette that accompanies this calculation.

<u>File Name</u>	<u>Description</u>
SRA012.DOC	This calculation in MS-WORD format
ATM11A.XLS	50%-ile Atmospheric Release Estimate for K-25 Site; EXCEL spread sheet
ATM11B.XLS	95%-ile Atmospheric Release Estimate for K-25 Site; EXCEL spread sheet
ENVIRO.XLS	Enrichment calculations for the environmental work; EXCEL spread sheet

Appendix A

SRA

August 22, 1996

Shonka Research Associates, Inc.

To: File

From: J. J. Shonka

Re: Task 6: Review of Release Fraction Literature

Memo No. JJS.048

cc T. Widner
J. Buddenbaum

A study of release fractions has been made. The release fractions under study relate to the fraction of uranium that would be released from a loss of process gas (UF_6) at a gaseous diffusion plant. Task 6 of the Oak Ridge Dose Reconstruction is charged with independently estimating the releases of uranium from the Oak Ridge Reservation, including the gaseous diffusion plant. The effort is directed towards establishing any errors in reporting that would cause significant changes in the site asserted releases. In the earlier feasibility study, the site asserted releases as documented in ORO-890 for the maximum year of releases was used as a screening estimate. Based on that estimate, the offsite impact of uranium releases was small compared to other significant contaminants of concern. Because uranium was used in large quantities at the Oak Ridge Reservation, this result seemed counter-intuitive to members of the review panel. The review panel suggested that in this phase of the project the sources of uncertainty and the potential for unmonitored releases be considered.

Many of the releases that form the basis of ORO-890 were not measured, but were asserted from accountability records of site operations. These records would assert that a release occurred at a given place and time and that a given quantity of Uranium was lost and presumed released. While not explicitly stated, either in ORO-890 or in its source documentation from the ORGDP, the previous source terms assumed that virtually all of the material was released with none of it held up on the surfaces of the plant and other systems. This would imply a large value, approaching 100% was used as a release fraction. This memorandum summarizes available scientific literature on UF_6 releases, and concurs that a 100% release fraction is possible and appropriate given the uncertainty in the size of release and other parameters.

Discussions were held with LMES technical staff concerning UF_6 behavior in a release. This behavior is qualitatively summarized as follows: for a release of UF_6 to occur, the source material must be at elevated temperatures and pressures relative to ambient conditions. At ambient temperatures, UF_6 is a solid. Under process diffusion plant conditions, the released UF_6 forms a white cloud from reactions with atmospheric moisture in air, forming hydrated uranyl fluoride ($UO_2F_2 \cdot H_2O$) and hydrofluoric acid (HF). The reactions are exothermic and, combined with the process thermal energy present inside of a gaseous diffusion plant, result in the rapid ascent of the white cloud of reaction products to the ceiling. (For ground level releases with pressures and temperatures just above the triple point, the cloud may remain near the ground.) The ceiling of cells of a gaseous diffusion plant are provided with periodic vents. The size and spacing of the vents is designed for removal of the hot air given off by process equipment, and not necessarily for removal of noxious contaminants. If the release occurs immediately below a vent, essentially all of the released material will exhaust through the vent. If the release occurs at some distance from a roof vent, the material rises to the ceiling and spreads out, gradually forming particulate (which partially deposits on surfaces) while the material migrates to and is entrained into nearby vent exhausts. In most cases, nearly all of the uranium is exhausted from the plant unless special measures are taken to limit the release (close all vents and inject steam into the cell to fully react the UF_6 and provide condensation nuclei to enhance fallout, has been attempted for example). Thus, a release fraction of 100% is appropriate and does not offer a substantial degree of conservatism, if any. Supporting evidence for this assumption is the lack of significant fallout found in several release tests of UF_6 (both indoors and outdoors) to the environment.

One of the most applicable reports is K-GD-916, "Containment of Released Uranium Hexafluoride" by R. L. Ritter (11/7/73). This report, as reviewed by the analyst, is an unclassified, redacted version of a short report of 1973 release studies in Cell K-902-5.9. Fifteen minute releases at rates from 10 to 100 grams UF_6 per minute were visually observed by a window placed into the cell wall, with air concentrations measured inside and outside the cell. Deposition studies were made using one square foot fallout pans which were placed inside and outside the cell and the release fraction to outside the building was estimated. UF_6 in moist air at room temperatures rapidly forms a white cloud. Because of elevated temperatures ($>160^\circ F$) in the cell as well as other differences (such as small release rate and high air flow rates) this white cloud was never observed either in the cell, building or outdoors. The relative humidity outdoors was likely low as well, given the time of year. Thus, heating the outdoor air to an ambient temperature of more than $160^\circ F$ would also produce in-cell air at a low relative humidity. A haze was observed sometime after the larger of the releases began. The white cloud was observed when the releases were made outside of the cells, although that portion of the report remains classified. Typical release fractions to the environment were asserted to be between 20% and 40%, although one of six experiments where air concentrations were measured had a

significantly lower release fraction of 5.3% that could not be explained by the authors. If you remove the low value, the average release fraction measured was 27% +/- 7% (one sigma). The cells had background deposition rates of two to four $\mu\text{g U/sq.ft./24 hours}$. Negligible fallout rates were observed above background inside the cell for release rates of less than 50 g/min. At higher release rates, slight fallout was observed in the cell and on the roof of the building. There is a roof vent immediately above the cell where the experiments were conducted. An average of 900 linear feet per minute was observed with high variability over the 32 square feet of vent area for an assumed 28,800 CFM flow rate. On three of the runs, the louvers were in the 10% open position which caused even greater air flow variations. The measurements of concentration across the varying air flow in the vent made asserting a release fraction difficult, and the author suggested that his data was more qualitative than quantitative.

The release fractions asserted by the author were based on a dynamic experiment that was difficult to control. The release would mix into the cell volume, which could likely be modeled by a first order linear kinetics model ($\exp(-\mu t)$) which would have an exponential response (with a time constant consistent with the cell air changes per hour) to a step change in input rate (e.g. from 0 to 50 grams per minute). The cell concentration was likely not at equilibrium until the release was terminated and the residual contents of the cell equilibrated (if they ever did). The cell contents, which are time varying, would mix, perhaps imperfectly with the access tunnel ventilation flow rate and be ejected out the vent. The vents were about 25 meters above the ground, and heat and ventilation cause the plume to be ejected another 15 meters up, for an effective release height of about 50 meters. The flow rate across the ventilation system was non-uniform, the air concentrations were non-uniform, and the releases were only maintained for 15 minutes into the cell, which would be smeared out in time by the mixing into the cell volume and exhaust. Thus, the observation by the author that the release fraction data should be viewed as qualitative are appropriate. The data was not analyzed for mass balance. A primitive mass balance could be derived from the fact that even though the largest releases had between 1200 and 1500 grams released, only tens of micrograms per square foot were observed as fallout. With cell areas of substantially less than millions of square feet, fallout accounted for less than one gram of material or at most a few grams) over 24 hours. Where did 99.9% of the material go? It had to either accumulate in an undiscovered area of the process building, or it had to go out with the airflow. Thus, although the analysis emphasized the air concentration, this was a dynamic variable that was difficult to measure. When one simply considers the lack of fallout of the material, release fractions of 99% or more cannot be eliminated and are consistent with the data. The applicability to releases to moist air at lower temperatures is not entirely straightforward. Clearly, the reaction rates were suppressed due to lack of atmospheric moisture. Thus, the formation of particulate would occur at a reduced rate, perhaps suppressing the amount of uranium that would appear as plate-out. The lack of equilibrium in the measurements

would be one reason for the data to be biased to low release fractions. Additionally, the ratio of the sampler flow rate to the vent flow rate is subject to considerable uncertainty, providing another potential source of bias. Finally, there is potential for a fraction of the release to exhaust from distant vents, since the air containing the reacting UF_6 would rise to the ceiling of the cell.

Report K-D-1894, "ORGDP Container Test and Development Program Fire Tests of UF_6 -Filled Cylinders" by A. J. Mallett (1/12/66) reported the destructive testing of potential shipping containers to fire environments during October of 1965. The tests were destructive, with two cylinders and three capacities of 5, 55 and 250 pounds of UF_6 used at an enrichment of 0.22%. The tests were primarily designed to observe the cylinder behavior in a fire. Of the data taken, the lack of significant air concentrations and fallout were noted. This report provides additional justification that the behavior of plumes of UF_6 is not affected in a significant fashion by the relatively high mass of the molecule, and the plume behaves in a manner similar to other chemical fire plumes.

Report KY/L-1213, "Assessment of Consolidated UF_6 Release Studies" by D. E. Boyd, C. G. Jones, and S. F. Seltzer (9/7/83) reported the efforts by DOE to consolidate UF_6 studies at the various plants to avoid duplication of effort. The report summarizes the studies each of the plants was conducting and was primarily a source of references on the related work. Work summarized from the Paducah Gaseous Diffusion Plant's KY/L-725 report "UO₂F₂ Particle Size Analysis by the Coulter Counter Method" indicated that the measured UO₂F₂ particle size ranged from 0.8u to 40u with the predominant size in the 0.8u to 2.5u range.

Report KY-795 "Fallout of Uranium During UF_6 Releases (UU)" is a 1/6/94 report by T. J. Mayo that summarizes data from two reports written in 1975 (KY/L-694 and KY/L-765). These reports discussed experiments where a heated bulb was charged with 215 grams of UF_6 and 14 grams of SF_6 at 58 psia. The contents were allowed to escape in the field, which resulted in the release of 160 grams of UF_6 and 10 grams of SF_6 . Eight releases were characterized in the first report, four in the second report. It was necessary to perform the releases late in the day to avoid excessive atmospheric dispersion which resulted in experimental data below the detection limit of the experiments. A chemically treated filter paper was used to measure the HF and UO₂F₂ as a function of distance. Measurements were made to distances of up to 400 meters, with small quantities of uranium observed. The author argued that the observations appear to support a conclusion that one cannot assume a large fraction of the uranium will quickly fallout from a cloud of reacting UF_6 .

Between 2/25/76 and 8/17/76 a total of 57 test releases involving a total of 2032.5 grams of UF_6 were made in K-33. The need for the tests was driven by the change in the K-33 ventilation system. The project has a cover letter concerning this experiment and is attempting to recover this document.

K/D-6092 Analysis of the June 5, 1989, UF₆ Release Test (2/93) by S. G. Bloom is an analysis of a cooperative release study performed in France. A series of UF₆ release tests were conducted by the French at their government test site at Bordeaux, France. About 150 Kg of UF₆ was released over 30 minutes at a height of 3 meters. Information included meteorological data, uranium and fluorine concentrations, particle size distribution, deposition data and visual observations. The US interest was in developing the data to benchmark an environmental transport code for UF₆ that accounted for chemical and physical transformations in a chemically reacting plume. The data showed a small particle size distribution for uranium that experienced deposition velocities from 0.01 to 2 centimeters per second, with an Andersen Impact Sampler measure mean particle size of about 3 μmeters. The data showed decreasing deposition with distance.

Finally, Report KY-L-824, "The Application of the Gaussian Plume Model Equation to UF₆ Releases" by T. J. Mayo (4/15/76) reported both SF₆ releases and studies of uranium fallout at the Paducah Gaseous Diffusion Plant. The conclusions of the report were that fallout would not be a major factor in reducing uranium concentrations in air at least to distances of several hundred meters.

Note added 11/1/96:

Earlier drafts of this memorandum were provided to staff (B. Manninen) at an operating gaseous diffusion plant (Portsmouth) for review and comment (See JJS.049). Telephone conversations were later held to obtain their comments and reactions to the assertions of release fraction. Prior to this memorandum, the staff felt that releases immediately below vents would have near total release fractions, but that would not occur for releases from equipment located some distance from a vent. In the discussions held after they had reviewed this data, their position changed to one which agreed with this memo, largely because of the particle size distribution observed in the French experiments (K/D-6092).

MEMORANDUM

Date: 10/23/96

To: Joe Shonka

From: Jack Buddenbaum

cc: Tom Widner, Jennifer Cockroft

Subject: 1986 - 1995 K-25 Uranium Airborne Releases

I have summarized below K-25 air release estimates for 1986 - 1994. The 1995 numbers can be obtained from a web site identified below. I have also attached to this memo, three FOIA incident notification summaries from the National Response Center. These reports describe K-25 uranium releases that may have not been included in Task 6 estimates. Let me know what you think.

K-25 Atmospheric Releases for 1986 - 1996

<u>Year</u>	<u>Curies</u>	<u>Kilograms</u>
1986	0.001	0.196
1987	0.0003	0.4
1988	0.0011	1.71
1989	0.0004	1.11
1990	0.0011	2.01
1991	0.0240	40.22
1992	0.0640	112.39
1993	0.01	12
1994	0.008	10
1995	*	*

* - The Oak Ridge Reservation Annual Site Environmental Report for 1995 can be retrieved from http://www.ornl.gov/Env_Rpt/aserep/asere.htm.

Let me know if you can retrieve the 1995 K-25 releases numbers from this web site. I can retrieve them here as soon as our IT coordinator returns to the office. Please cc-mail to me ASAP the updated spreadsheet that includes 1944 - 1995 atmospheric releases for K-25.

Let me know if you have questions or comments.

Thanks,

Jack

NATIONAL RESPONSE CENTER

*** FOIA INCIDENT REPORT 7179 ***
FOR 1985

INCIDENT DESCRIPTION

Report taken by BROWN on 31 at 1614
Incident type: F
Affected Medium: ATMOS
The incident occurred on 30 JUL85 at 1730 local time.
Weather: N Sea: N Current: N
Color: N Wind: N
Sheen Size: N
Vessel/Vehicle:
Consignee:

SOURCE/CAUSE OF INCIDENT
RESIDUAL IN LINE BLOWN OUT STACK

INCIDENT LOCATION

OAKRIDGE GASIOUS DIFFUSION PLANT BLDG K-31 HWY 58 OADRIDGE
TN ROAN CNTY

RELEASED MATERIAL(S)

CHRIS Code: MIS URANIUM HEXAFLORIDE (GAS) RAD
Qty Released: 2.2 LBS
Qty in Water: N

DAMAGE

Injuries: 0 Fatalities: 0
Damages:

REMEDIAL ACTIONS

NONE

]

REPORTING PARTY

Organization: DOE
Address: BX E OAKRIDGE TN 37831
State: TN

Calling for Responsible Party: Y

NOTIFICATIONS

B

EPA AND STATE OF TN

ADDITIONAL INFORMATION

*** END FOIA INCIDENT REPORT 7179 ***

NATIONAL RESPONSE CENTER

*** FOIA INCIDENT REPORT 16444 ***
FOR 1988

INCIDENT DESCRIPTION

Report taken by RCP on 25-SEP-88 at 1646

Incident type: F

The incident occurred on 25-SEP-88 at 1525 local time.

Vessel/Vehicle:

SOURCE/CAUSE OF INCIDENT

SPILL FROM A MIXING TRUCK. DUE TO OPERATOR ERROR.
TRUCK WAS PARKED AT THE FACILITY

INCIDENT LOCATION

NEAR HWY 95

County: ROAN

City: OAKRIDGE

St: TN

RELEASED MATERIAL(S)

Chris

Code: Material Name:

Total Qty: Units: In Water: Units:

RAM URANIUM/F006

10.00 LBS 0.00 NON

0.00

0.00

0.00

0.00

DAMAGE

Injuries:

Fatalities: Evacuations: 0

Damages: 0

Amount:

REMEDIAL ACTIONS

MATERIAL WAS PICKED UP AND PUT BACK INTO THE MIXER
CLEANED.

REPORTING PARTY

Organization: DEPT OF ENERGY OAKRIDGE GASEOUS

Addr:

State: TN Zip: 37831-

Calling for Responsible Party: 1

SUSPECTED RESPONSIBLE PARTY

Organization:

Addr:

State: Zip:

NOTIFICATIONS

EPA Region: 4 Time: 1703

MSO/COTP: Time: ?

Caller Notified: TN EMER MGT

Others Notified: NRC-1702, DOE-1707

ADDITIONAL INFORMATION

*** END FOIA INCIDENT REPORT 16444 ***

*** FOLA INCIDENT REPORT 16284 ***
FOR 1988

Report taken by AKL on 21-SEP-88 at 2356
Incident type: F
The incident occurred on 21-SEP-88 at 2150 local time.
Vessel/Vehicle:

LARGE DRUM/ FELL OFF PALLET AND BROKE

OAKRIDGE GASEOUS DIFFUSION PLANT

County: ROANE **City:** OAKRIDGE **St:** TN

Chris				
Code:	Material Name:	Total Qty:	Units:	In Water: Units:
NCC	URANIUM/F006	1500.00	LBS	0.00 NON
	0.00	0.00		
	0.00	0.00		

Injuries: Fatalities: Evacuations: 0
Damages: 0 Amount:

SPILL HAS BEEN CONTAINED AND IS BEING CLEANED UP.

Organization: DEPT OF ENERGY, OAKRIDGE, TN
Addr: FEDERAL OFFICE BUILDING
State: TN Zip: 37831-

Calling for Responsible Party: I

Organization:
Addr:
State: Zip:

NOTIFICATIONS

EPA Region: 4 Time: 24

MSO/COTP: Time: ?

Caller Notified: TN EM

Others Notified:

ADDITIONAL INFORMATION

*** END FOIA INCIDENT REPORT 16284 ***

SHONKA RESEARCH ASSOCIATES, INC.

Calculation Control Sheet

Calculation number: SRA-95-010 REV. 0Title: URANIUM RELEASE ESTIMATES FOR THE ORG-DIP PURGE
CASCADE 12/45-12/46Reason for calculation/revision: ORIGINAL
✓Client: CHEMRISK, INC.Project: BAK RIDGE HEALTH STUDIESProject/Task Number: TASK 6Prepared by: TE BENNETTDate: 11/9/95

Post-it® Fax Note		7671	Date	# of pages
To			From	
Co./Dept.			Co.	
Phone #			Phone #	
Fax #			Fax #	

Independent Technical Review by: Joseph J. ShonkaDate: 12/1/95Quality Assurance Review by: Dalnah B. ShonkaDate: 12/4/95
☐ This calculation has been voided or superseded by _____
(calculation number)

This document has been reviewed for classification and has been determined to be UNCLASSIFIED.	
<u>Thomas W. Kelly</u>	
ADC Signature	
<u>12/7/95</u>	
Date	

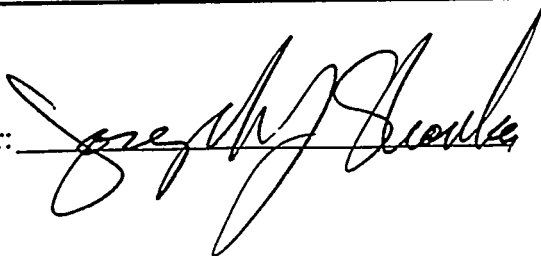
SHONKA RESEARCH ASSOCIATES, INC.

Review Method Sheet

The undersigned has reviewed this calculation in accordance with the method(s) indicated below.

1. Computer Aided Calculation	
a	Review to determine that the computer program(s) has been validated and documented, is suitable to the problem being analyzed, and that the calculation contains all necessary information for reconstruction at a later date.
b	Review to determine that the input data as specified for program execution is consistent with the design input, correctly defines the problem for the computer algorithm and is sufficiently accurate to produce results within any numerical limitations of the program.
c	Review to verify that the results obtained from the program are correct and within stated assumptions and limitations of the program and are consistent with the input.
d	Review validation documentation for temporary changes to listed, or developmental, or unique single application programs, to assure that the methods used adequately validate the program for the intended application.
e	Review of code input only, since the computer program has sufficient history of use at Shonka Research Associates, Inc. in similar calculations.
f	Review arithmetic necessary to prepare code input data.
g	Other:
2. Hand Prepared Calculations	
<input checked="" type="checkbox"/> a	Detailed review of the original calculations.
<input checked="" type="checkbox"/> b	Review by an alternate, simplified, or approximate method of calculation.
c	Review of a representative sample of repetitive calculations.
d	Review of the calculation against a similar calculation previously performed.
e	Other:
3. Revisions	
a	Editorial changes only
b	Elimination of unapproved input data without altering calculated results.
c	Other:
4. Other	

Reviewer:



Date:

12/1/95

CALC NO SRA-95-010 REV 0Project/Task ChemRisk/TDH Oak Ridge Dose ReconstructionPrepared by T. E. BennettDate 11/8/95

Checked by/Date

J. J. ShonkaTitle Uranium Release Estimates for the ORGDP Purge Cascade 12/45-12/46 (signed original on file)

ABSTRACT

This calculation represents the second in a series of calculations aimed at establishing a methodology and strategy in analyzing the quality of the historical effluent monitoring data and practices at the Oak Ridge Gaseous Diffusion Plant. Within that scope, the emission rates from the purge cascade during the months of December 1945 through December 1946 were analyzed in order to determine its magnitude as a site contributor in the total historical uranium emissions. Daily purge rate data that documented the volumetric flow of the purge gas and its concentration of UF_6 were used to compute the daily flow of UF_6 in the purge cascade. The total volumetric flow of UF_6 for each month was used in the modified van der Waals real gas equation in order to estimate the mass of UF_6 released. Calculations show that over the thirteen month period, about 75.9 g of moderately enriched UF_6 was measured by the space recorder monitoring instrumentation located in cells K-312-1, K-312-2, and K-312-3 of the purge cascade. This corresponds to the release of about 916 μCi of alpha activity and includes contributions from the mixture of $^{234}\text{UF}_6$, $^{235}\text{UF}_6$, and $^{238}\text{UF}_6$. These estimates do not include any reductions due to carbon or alumina traps located downstream of the monitoring instrumentation prior to the purge stack exit.

CALC NO SRA-95-010 REV 0

Project/Task ChemRisk/TDH Oak Ridge Dose Reconstruction

Prepared by T. E. Bennett

Date 11/8/95

Checked by/Date

J. J. Shonka

Title Uranium Release Estimates for the ORGDP Purge Cascade 12/45-12/46 (signed original on file)

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Attachments

1. Purge Rates November 1945 (partial) - December 1946 (from the K-25 Process Division Daily Reports at K-1034-A Site Records)

CALC NO SRA-95-010 REV 0Project/Task ChemRisk/TDH Oak Ridge Dose ReconstructionPrepared by T. E. BennettDate 11/8/95

Checked by/Date

12/1/95J. J. ShonkaTitle Uranium Release Estimates for the ORGDP Purge Cascade 12/45-12/46 (signed original on file)

1. INTRODUCTION

Task 6 of the Oak Ridge Dose Reconstruction focuses on the evaluation of the quality of historical airborne and waterborne effluent monitoring data and the determination of the potential significance of unmonitored emissions. Uranium played an important role throughout historical operations on the Oak Ridge Reservation (ORR) and is known to have been released to the environment through air and water. The two largest uses of uranium on the Reservation were the enrichment processes of the ^{235}U isotope by electromagnetic separation at the Y-12 facility and gaseous diffusion at the K-25 facility.

Task 6 activities will be directed at establishing revised uranium release estimates with an associated uncertainty over that of the screening analyses conducted during the Dose Reconstruction Feasibility Study. These activities will support refined assessment of the potential magnitude of health hazards from historical uranium exposures based on both the chemical and radiotoxicity of uranium.

This calculation represents the second in a series of calculations aimed at establishing a methodology and strategy in analyzing the quality of the historical effluent monitoring data and practices at the Oak Ridge Gaseous Diffusion Plant (ORGDP). For many years the purge cascade represented the only on-site monitored emission source. This along with previous and subsequent calculations will provide a historical review of the uranium released from the ORGDP purge cascade including an assessment of the potential uncertainties and biases in the measurement and computation of the estimates.

Light molecular weight gases were purged from the top of the diffusion cascade that would otherwise block the withdrawal of enriched UF_6 product. These light gases originated from the following sources.

- *nitrogen* - mainly from the inleakage occurring at every pump shaft seal in the diffusion cascade
- *hydrogen fluoride* - from reaction of inleaking moist air with UF_6
- *oxygen, argon* - from inleaking air
- *chlorine fluorides* - used in conditioning and drying of metal surfaces
- *fluorine* - used in conditioning of metal surfaces
- *coolant vapor* - inleakage from the compressor and pump coolant system

The light gases in the process stream had a molecular weight substantially less than that of the UF_6 component and were carried along effectively by the diffusion process to the top of the cascade. A section at the top of the cascade just above the product withdrawal point was reserved as a purge cascade. The purge cascade separated these light gases from the enriched UF_6 product and vented them to the atmosphere. The purge cascade usually existed in two sections: a side purge and a top

CALC NO SRA-95-010 REV 0Project/Task ChemRisk/TDH Oak Ridge Dose ReconstructionPrepared by T. E. Bennett Date 11/8/95 Checked by/Date J. J. Shonka JSTitle Uranium Release Estimates for the ORGDP Purge Cascade 12/45-12/46 (signed original on file)

purge. The side purge separated the intermediate molecular weight gases (*i.e.* coolant vapor, chlorine fluorides, etc.) from the UF_6 . The top purge separated the remaining lighter gases. At various times throughout the operational history, the side purge was routed to the top purge. The effluents were pumped through traps in order to reduce the uranium content before monitoring and venting to the atmosphere. In the earlier years these traps consisted only of carbon and alumina, but later sodium fluoride (NaF) traps and potassium hydroxide (KOH) scrubbers were added. Because of the large difference in molecular weights between the light gases and UF_6 , only a few diffusion stages were needed to effectively perform the separation. Similar to the main diffusion cascade, the purge cascade consisted of compressors, converters, motors, coolers, piping, control and block valves, and instrumentation. The major difference between the main diffusion cascade and the purge cascade is the smaller amount of UF_6 flow (MMES 1985).

The problem of analyzing the gas in the purge cascade was complicated by the fact that the UF_6 concentration varied greatly from one end of the cascade to the other. Near the bottom of the cascade the process stream consisted of essentially pure UF_6 , whereas at the cascade top the stream consisted of light gases containing only small traces of UF_6 . The line recorder was designed for analyzing UF_6 containing relatively small amounts of impurities. The method it employed measured the flow of gas to a mass spectrometer tube by means of a Pirani gauge flowmeter; the UF_6 was removed chemically before reaching the spectrometer tube and the residual gas concentration measured by means of the spectrometer tube. In analyzing for UF_6 in the presence of relatively large amounts of impurities, the accuracy dropped sharply. Due to the corrosive nature of UF_6 , the concentration could not be directly measured by the spectrometer tube. Therefore, the UF_6 concentration was determined by the difference between the flow computed from the flowmeter and the remaining light gas concentration determined by the mass spectrometer tube reading. Only with careful calibration was it possible to determine either of these quantities with an accuracy of 1% or better. Accordingly, the line recorder became practically useless for determining the composition of a mixture containing under 2% of UF_6 (OM-48 1945).

In order to supplement the line recorder in the purge cascade, an instrument known as the space recorder was developed. The principal component of the space recorder was an ionization chamber more commonly referred to as the "signal can." The signal can measures the specific radioactivity of the gas present, and since UF_6 is an alpha emitter, this method provided a convenient means for measuring the UF_6 content of gas samples. The space recorder could detect the presence of mol fractions of UF_6 in the light gas purge of the order of 10^{-6} (OM-48 1945). The radioactivity of UF_6 consists of the emission of high energy alpha particles at a definite uniform rate. This rate depends upon the relative isotopic composition, since all three isotopes emit alpha particles at a different rate. These alpha particles as emitted have a definite range of travel, which is inversely proportional to the pressure. In the gas samples, at standard temperature and pressure, this range of travel is on average approximately 3 cm. While travelling this distance the energy of the emitted particle is expended by the production of about 130,000 ions resulting from collision of the particles with gas molecules.

CALC NO SRA-95-010 REV 0

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Prepared by T. E. Bennett

Date 11/8/95

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Title Uranium Release Estimates for the ORGDP Purge Cascade 12/45-12/46 (signed original on file)

The collection of these ions and a measurement of the resultant electrical current constitutes a determination of the number of particles present and hence a determination of the UF_6 concentration. A record of the purge gas volumetric flow and the concentration of UF_6 in the purge stream during the 1945-1946 time frame was reported in the Process Division daily reports.

This calculation analyzes a thirteen month span of daily purge cascade effluent data for the months of December 1945 through December 1946. These estimates do not represent an actual estimation of the amount of uranium released to the atmosphere during this time period. The sample withdrawal point for the space recorders were located upstream of the carbon and alumina traps. The traps would have removed much of the uranium prior to the venting of the purge gases to the atmosphere and these benefits are not included in these estimates. With this limitation in mind, this calculation makes liberal use of the terms "release", "released", "vented", and "effluent." Furthermore, these estimations are also subject to certain assumptions, biases, and other uncertainties. Several assumptions and potential sources of uncertainty will be presented in this calculation, but the analyses to quantify the impact of these assumptions to the results of this calculation will be formalized in subsequent calculations.

In this calculation, data sheets containing the daily purge rates for the months of December 1945 through December 1946 were transcribed from the Process Division daily reports and transferred to spreadsheets (Purge Rates 1946). The volume of gas purged each day and its UF_6 concentration was used to compute the daily volumetric flow of UF_6 released. The daily flow of UF_6 was summed to compute the estimate of the total volume of UF_6 vented during the month. The mass of UF_6 released each month in the purge cascade was derived from this known volume at standard conditions by application of the modified van der Waals real gas equation (Ackley, Magnuson 1951) as given in Eq. 1-1.

$$m = \frac{P(1 + AP)V}{RT} \quad (1-1)$$

where P is the pressure of the gas,

A is the temperature-dependent van der Waals coefficient for UF_6 ,

V is the volume of the gas,

R is the UF_6 gas constant, and

T is the temperature of the gas.

The activity of UF_6 released each month in the purge cascade was computed by multiplying the grams of UF_6 by the specific activity of UF_6 at the assumed ^{235}U enrichment level. The "effective" specific activity of a mixture of $^{234}\text{UF}_6$, $^{235}\text{UF}_6$, and $^{238}\text{UF}_6$ (as found in the purge cascade effluent) follows Eq. 1-2.

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$$S = (0.4 + 0.38E + 0.0034E^2) \times 10^{-6} \text{ Ci/g} \quad (1-2)$$

where E is the percent ^{235}U by weight. Eq. 1-2 is fitted to the experimental data in Fig 1-1 (Rich 1988). The contribution to the total "effective" activity of each isotope of uranium was determined from the graph presented in Fig 1-2 (Rich 1988) and used to determine the activity of each isotope. The mass of ^{234}U , ^{235}U , and ^{238}U in the purge effluent could then be calculated from its activity and theoretical specific activity as given by Eq. 1-3. The results of the isotopic mass calculations were compared to the mass calculations for UF_6 using Eq. 1-1 in order to determine the "goodness" of the values selected from Fig 1-2.

$$m_i = \frac{A_i}{S_i} \quad (1-3)$$

where A_i is the activity of the radioisotope, and
 S_i is the specific activity of the radioisotope.

The theoretical specific activity of each uranium isotope is calculated by Eq. 1-4.

$$S_i = \frac{\lambda_i N_A}{M_i} \quad (1-4)$$

where λ_i is the decay constant of the radioisotope,
 N_A is Avogadro's Number, and
 M_i is the atomic weight of the radioisotope.

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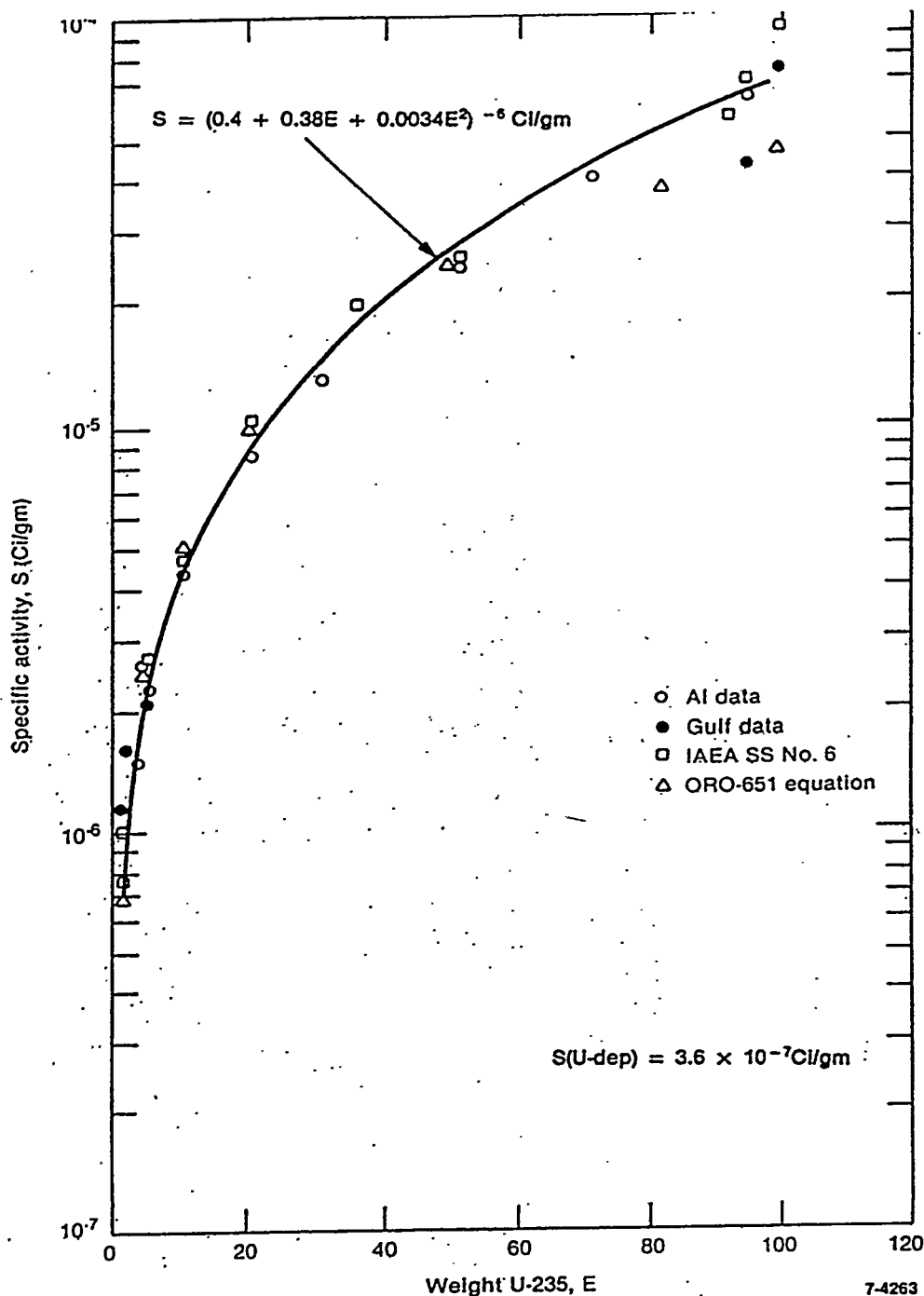


Fig 1- 1 : Specific Activity for Mixtures of ^{234}U , ^{235}U , and ^{238}U

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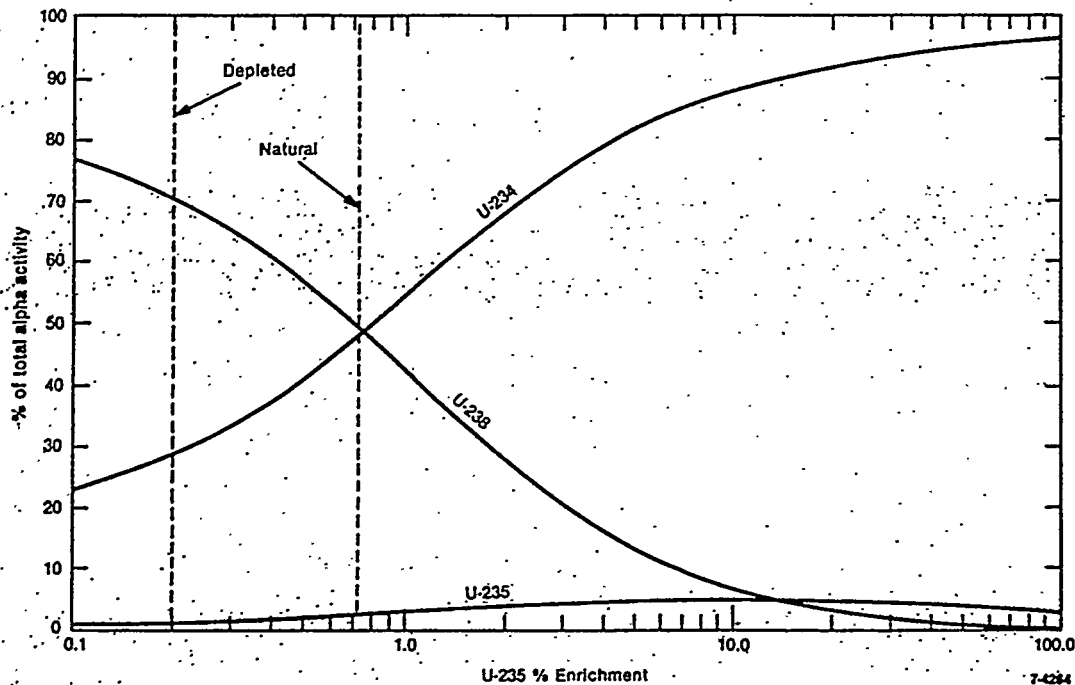


Fig 1-2 : Percent of total radioactivity by isotope vs. % weight ^{235}U enrichment

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2. SUMMARY OF RESULTS

The monthly uranium release estimates from the ORGDP purge unit between December 1945 and December 1946 are presented below in Table 2-1. Totals are given for the sum of the thirteen month period. In addition, the median and standard deviation are given for all computations. The first column of data presents the results of the UF_6 mass calculations using the modified van der Waals real gas equation in Eq. 1-1. The mass of the F_6 is subtracted from the UF_6 mass and the results presented in the second column along with the computation of the "effective" activity given by Eq. 1-2. The next three columns of data present the mass and activity of ^{238}U , ^{235}U , and ^{234}U respectively. The activity of each of the three constituents are computed by applying the appropriate activity fraction from Fig 1-2 to the "effective" activity. The mass of each of the three constituents are computed using the theoretical specific activities of each radioisotope. The last column presents the sum of the masses of each of the three constituents and the percent difference with the uranium mass based upon the modified van der Waals real gas equation computation.

The total UF_6 release over the thirteen month period is 75.94 g with 51.28 g being uranium. This corresponds to a total "effective" activity of 916.1 μCi and assumes a 35% ^{235}U enrichment. The "effective" activity corresponds to alpha decay contributions from the mixture of ^{234}U , ^{235}U , and ^{238}U . According to Fig 1-2 at 35% ^{235}U enrichment, virtually all of the activity is due to the presence of ^{234}U . At this assumed enrichment, about 94.5% of the activity results from the ^{234}U isotope, 4.3% from the ^{235}U isotope, and the remaining from the ^{238}U isotope. The results also indicate that at this assumed ^{235}U enrichment, the ^{234}U isotope is enriched from 0.0056% natural abundance to just over 0.27% and the ^{238}U isotope is depleted to under 64%. The sum of the three isotopic masses understate the calculated uranium mass from the van der Waals gas equation by 0.36%. This difference is subject to uncertainty in the experimental data and the fitted equation in Fig 1-1 and to uncertainty in reading the data from the graph in Fig 1-2.

These estimates do not include any reductions due to carbon or alumina traps located downstream of the space recorder instrumentation prior to the purge stack exit.

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Table 2-1: Purge Cascade Uranium Release Estimates Dec 1945-Dec 1946

Month	UF ₆	U		U-238		U-235		U-234		Total	
	[g]	[g]	[Ci]	[g]	[Ci]	[g]	[Ci]	[g]	[Ci]	[g]	Δ%
Dec-45	9.381E+00	6.334E+00	1.132E-04	4.042E+00	1.358E-06	2.252E+00	4.866E-06	1.722E-02	1.069E-04	6.311E+00	-0.36%
Jan-46	2.738E+01	1.849E+01	3.302E-04	1.180E+01	3.963E-06	6.573E+00	1.420E-05	5.026E-02	3.121E-04	1.842E+01	-0.36%
Feb-46	1.845E+01	1.246E+01	2.226E-04	7.950E+00	2.671E-06	4.430E+00	9.571E-06	3.387E-02	2.103E-04	1.241E+01	-0.36%
Mar-46	8.637E+00	5.832E+00	1.042E-04	3.721E+00	1.250E-06	2.074E+00	4.480E-06	1.586E-02	9.845E-05	5.811E+00	-0.36%
Apr-46	1.634E+00	1.103E+00	1.971E-05	7.040E-01	2.365E-07	3.923E-01	8.475E-07	3.000E-03	1.862E-05	1.099E+00	-0.36%
May-46	7.719E+00	5.212E+00	9.311E-05	3.326E+00	1.117E-06	1.853E+00	4.004E-06	1.417E-02	8.799E-05	5.193E+00	-0.36%
Jun-46	3.300E-01	2.228E-01	3.981E-06	1.422E-01	4.777E-08	7.924E-02	1.712E-07	6.059E-04	3.762E-06	2.220E-01	-0.36%
Jul-46	4.114E-01	2.778E-01	4.962E-06	1.773E-01	5.955E-08	9.877E-02	2.134E-07	7.552E-04	4.690E-06	2.768E-01	-0.36%
Aug-46	3.674E-01	2.481E-01	4.432E-06	1.583E-01	5.318E-08	8.821E-02	1.906E-07	6.745E-04	4.188E-06	2.472E-01	-0.36%
Sep-46	3.869E-01	2.612E-01	4.667E-06	1.667E-01	5.600E-08	9.289E-02	2.007E-07	7.103E-04	4.410E-06	2.603E-01	-0.36%
Oct-46	3.873E-01	2.615E-01	4.672E-06	1.669E-01	5.606E-08	9.298E-02	2.009E-07	7.110E-04	4.415E-06	2.606E-01	-0.36%
Nov-46	3.965E-01	2.677E-01	4.783E-06	1.708E-01	5.739E-08	9.519E-02	2.057E-07	7.279E-04	4.520E-06	2.667E-01	-0.36%
Dec-46	4.667E-01	3.151E-01	5.629E-06	2.011E-01	6.755E-08	1.120E-01	2.421E-07	8.567E-04	5.320E-06	3.140E-01	-0.36%
Total	7.594E+01	5.128E+01	9.161E-04	3.272E+01	1.099E-05	1.823E+01	3.939E-05	1.394E-01	8.657E-04	5.109E+01	-0.36%
Median	4.667E-01	3.151E-01	5.629E-06	2.011E-01	6.755E-08	1.120E-01	2.421E-07	8.567E-04	5.320E-06	3.140E-01	-0.36%
Std Dev	8.191E+00	5.531E+00	9.881E-05	3.529E+00	1.186E-06	1.967E+00	4.249E-06	1.504E-02	9.337E-05	5.511E+00	-0.36%

In June 1946, a study was conducted and determined that the average product loss in the purge cascade during February, March, and April 1946 was 2.2×10^{-5} lbs. UF₆ per day (Moore 1946). The purge data collected and analyzed in this study estimate approximately 14.5×10^{-5} lbs. UF₆ per day, nearly a seven-fold difference. The product losses reported in June 1946 imply the estimates included the reductions of the carbon and alumina traps. If the space recorder data have been analyzed similarly in both calculations, this would lead to the conclusion that the trapping efficiency for UF₆ in the purge cascade during the 1945-1946 time frame to be approximately 85%.

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3. METHODS

The results summarized in Table 2-1 were based upon data sheets generated from the K-25 Process Division daily reports. These data sheets contain the daily log of the purge unit volumetric flow and UF_6 concentration as measured at purge cells K-312-1, K-312-2 and K-312-3, and are included in this calculation as Attachment 1. As part of this calculation, these data sheets were recreated as individual spreadsheets in a Microsoft[®] Excel workbook and are provided electronically in the file 95010R0.XLS. In order to determine an estimate for the total amount of uranium released by the purge unit over this thirteen month period, a number of computations using the daily purge data were required.

The purge unit consisted of three cells: K-312-1, K-312-2, and K-312-3. At any one time, two of the cells were in operation and the remaining cell in standby. One cell operated as a top purge and another cell operated as a side purge. Any of the three cells could operate as a top or side purge. The purge gas flow from the two cells were summed to compute the total daily flow of purge gases as well as the total monthly flow. The monthly total was then divided by the number of days in the month to compute the average daily purge gas flow.

The daily UF_6 volumetric flow was calculated by multiplying the UF_6 mole weight per cent and the purge gas volumetric flow. The results of this calculation for both the purge cells were summed to compute the total daily UF_6 volumetric flow in the purge cascade. These daily UF_6 flows were summed to compute the total UF_6 volume released by the purge cascade for the month.

The mass of UF_6 released by the purge unit was calculated using Eq 1-1 and the monthly total volume of UF_6 , the modified van der Waals pressure, standard temperature, and the UF_6 gas constant. The UF_6 mass was multiplied by the ratio of the molecular weight of uranium to the molecular weight of UF_6 resulting in the computation of the uranium mass. The "effective" specific activity of uranium at the assumed 35% ^{235}U enrichment was calculated using Eq 1-2 and the result multiplied by the uranium mass to compute the "effective" activity of the uranium. The fractional contribution of this "effective" activity by ^{234}U , ^{235}U , and ^{238}U are obtained from the graph in Fig 1-2 and each multiplied by the "effective" activity to compute the estimated activity of each isotopic constituent. The theoretical specific activity of each constituent was calculated using Eq 1-4 and then divided into the respective activity estimates according to Eq 1-3 to compute the estimated mass of each isotopic constituent. The three masses of the constituents were summed and compared to the uranium mass based upon Eq 1-1 in order to ensure the activity fractions obtained from the graph in Fig 1-2 were appropriate. The activity fraction values were iteratively refined until good agreement between the two mass calculations were obtained.

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4. ASSUMPTIONS

- 4.1. The atomic weight of ^{238}U is given as 238.05077 g/mol (Physics 1967).
- 4.2. The half-life of ^{238}U is given as 4.46×10^9 yr (Physics 1967).
- 4.3. The atomic weight of ^{235}U is given as 235.043915 g/mol (Physics 1967).
- 4.4. The half-life of ^{235}U is given as 7.04×10^8 yr (Physics 1967).
- 4.5. The atomic weight of ^{234}U is given as 234.040904 g/mol (Physics 1967).
- 4.6. The half-life of ^{234}U is given as 2.46×10^5 yr (Physics 1967).
- 4.7. The atomic weight of fluorine is given as 18.998403 g/mol (Physics 1967).
- 4.8. Avogadro's Number is given as 6.022045×10^{23} atoms/mol (Physics 1967).
- 4.9. For purposes of U and UF_6 molecular weight calculations, the contribution due to $^{234}\text{UF}_6$ are assumed negligible.
- 4.10. Standard pressure is assumed 14.7 psia (Lee 1989).
- 4.11. Standard temperature is assumed 59 F or 519 R (Lee 1989).
- 4.12. The universal gas constant \bar{R} is given as $10.73 \text{ psia ft}^3 \text{ lb}^{-1} \text{ mol}^{-1} \text{ R}^{-1}$ (Black, Hartley 1985).
- 4.13. The uranium enrichment of the UF_6 in the purge stream for the purposes of this calculation is assumed to be 35% ^{235}U . During the time frame of this calculation, the ORGDP was engaged in startup activities that limited the enrichment of uranium to ~35% ^{235}U . The precise enrichment characteristic of the product remains unknown.
- 4.14. The conversion factor of 453.6 g/lb is used to convert between units of mass (Black, Hartley 1985).
- 4.15. The conversion factors of 365.25 days/yr, 24 hr/day, and 3600 s/hr are used to convert between units of time.
- 4.16. The conversion factor of 3.7×10^{10} Bq/Ci is used to convert between units of activity.
- 4.17. The UF_6 concentrations recorded on the purge rate data sheets are assumed given as mole weight percentages.

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- 4.18. Purge gas flow and UF_6 concentration measurements in the K-312-1, K-312-2 and K-312-3 cells represent the sum total of the flow in the top purge unit.
- 4.19. This calculation assumes UF_6 in the purge stream behaves as a real gas following the behavior prescribed by the modified van der Waals gas equation. The van der Waals coefficient for UF_6 is a function of temperature and has the values of 0.033 atm^{-1} at 141.7 F and 0.021 atm^{-1} at 201.0 F (Ackley, Magnuson 1951).
- 4.20. The trap efficiencies are assumed negligible for the purposes of this calculation, thus implying that the purge gas flow and UF_6 concentration recorded on the purge rate data sheets are identical to the material actually vented to the atmosphere. The impact to the uranium release estimates due to actual trap efficiency, sampling biases and losses, and measurement uncertainties will be addressed in subsequent calculations. Some of the relevant sampling issues are (1) the maintenance of sufficient sample line temperature to prevent UF_6 condensation, (2) losses due to the geometry of sample lines, and (3) measurement uncertainties in the space recorder. The ^{235}U assay percent will also have an impact on the uranium release estimates.

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5. CALCULATION

5.1 Calculation of the UF_6 Gas Constant

The symbol R in Eq. 1-1 is known as the *gas constant* and its value depends upon the particular gas being considered. The value of R for each gas is determined by the equation

$$R = \frac{\bar{R}}{M} \quad (5.1-1)$$

where \bar{R} is a physical constant called the *universal gas constant* and is given in Section 4. It is first necessary to compute the atomic weight of uranium. Eq 5.1-2 gives the molecular weight of an isotopic mixture as

$$\frac{1}{M} = \frac{1}{100} \sum \frac{w_i}{M_i} \quad (5.1-2)$$

and for 35% ^{235}U

$$\frac{1}{M} = \frac{1}{100} \left(\frac{35}{235.043915} + \frac{65}{238.05077} \right),$$

which gives $M = 236.9896588$. The molecular weight of UF_6 is then $236.9896588 + (6)(18.998403) = 350.9800768$. Substituting into Eq. 5.1-1, the value of R for UF_6 becomes

$$R = \frac{10.73}{350.9800768} = 0.030571536 \text{ psia ft}^3 \text{ lb}^{-1} \text{ R}^{-1}.$$

5.2 Calculation of the Modified van der Waals Pressure

Since UF_6 behaves as a real gas, the modified van der Waals pressure is required to account for the non-ideality of the UF_6 in the gaseous diffusion process. The expression $P(1+AP)$ in Eq. 1-1 represents the modified pressure in the traditional ideal gas equation. The parameter A in the above expression represents the temperature-dependent van der Waals coefficient. Given values for the van

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der Waals coefficient at two temperatures, the following expression was derived that describes the nature of the van der Waals coefficient (atm^{-1}) as a function of UF_6 temperature (F).

$$A = -2.02 \times 10^{-4} T + 0.0617 \text{ atm}^{-1}$$

Naturally, as the temperature of the gas increases, the gas behaves in a more ideal manner. For standard conditions in which the temperature is given as 59 F, the van der Waals coefficient is given as $0.049782 \text{ atm}^{-1}$, or 0.0034 psia^{-1} . Using the van der Waals coefficient at standard conditions, the above expression evaluates the modified van der Waals pressure as

$$P' = 14.7 \times (1 + (0.0034)(14.7)) = 14.75 \text{ psia.}$$

5.3 Calculation of the Monthly UF_6 and Uranium Release Estimates

Table 5.1-1 depicts a sample daily purge rate data spreadsheet for the month of December 1945. The spreadsheets for the remaining months are in the Microsoft® Excel workbook 95010R0.XLS. The daily purge rates on the data sheets are given in units of standard cubic feet per day (scfd) and the Tops Conc are given units of mol wt % UF_6 .

For each day, the total purge gas volumetric flow and UF_6 concentration is calculated. The purge flow is the sum of the purge rate in the top (312-2) and side (312-3) purge units. The daily molecular weighted UF_6 percent in the purge gases is computed by multiplying the purge rate and the tops concentration and summing this value in both the 312-2 and 312-3 purge units. These daily rates are shown in the rightmost column in Table 5.1-1. For example, the per cent volume of UF_6 purged on December 7, 1945 would be calculated as

$$(1020 \times 0.00001) + (1395 \times 0.000012) = 0.02694 \% \text{scfd.}$$

The total volume of UF_6 purged for the month is the sum of these daily volumetric flows divided by 100. For December 1945, this computes to 0.022 ft^3 . The total volume of UF_6 at standard conditions is used in Eq. 1-1 to estimate the mass of UF_6 released from the purge unit for the month. The modified van der Waals pressure is substituted for the expression $P/(1+AP)$. This calculation for December 1945 precedes as follows

$$m = \frac{(14.75)(0.022)(453.6)}{(0.030571536)(519)} = 9.381 \text{ g } \text{UF}_6.$$

The uranium-only fraction of the UF_6 mass is calculated by multiplying by the ratio of the molecular weights of U and UF_6 computed in Section 5.1. For December 1945, the calculation precedes as

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$$m = \frac{(9.381)(236.9896588)}{350.9800768} = 6.334 \text{ g U.}$$

Table 5.1-1: Spreadsheet of Purge Rate Data for December 1945

Day of Month	312-2		312-3		Total	UF6 Purge
	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	
1	710	0.000032	1300	N/A	2010	0.02272
2	470	0.00004	1900	N/A	2370	0.0188
3	470	0.000025	1400	N/A	1870	0.01175
4	1100	0.00003	1800	0	2900	0.033
5	1210	0.000007	2360	0.000056	3570	0.14063
6	1187	0.000024	3065	0.00005	4252	0.181738
7	1020	0.00001	1395	0.000012	2415	0.02694
8	840	0.00001	1450	0	2290	0.0084
9	1125	0.00002	1435	0.00002	2560	0.0512
10	860	0.000136	1065	0	1925	0.11696
11	1160	0.00005	720	0.00001	1880	0.0652
12	680	0.00003	980	0.000025	1660	0.0449
13	385	0.000032	690	0.00004	1075	0.03992
14	1100	0.000011	900	0.000009	2000	0.0202
15	1040	0.0001	1020	0.00008	2060	0.1856
16	1360	0.000006	540	0.00142	1900	0.77496
17	1160	0.000015	1300	0.000004	2460	0.0226
18	1500	0.00004	990	0.00005	2490	0.1095
19	960	0.000002	840	0	1800	0.00192
20	970	0.000003	610	0.000056	1580	0.03707
21	1030	0.000002	1220	0.000035	2250	0.04476
22	1034	0.00003	2052	0.000031	3086	0.094632
23	947	0.000006	2465	0.00003	3412	0.079632
24	1108	0.000006	1455	0.000003	2563	0.011013
25	997	0.000003	1045	0.000003	2042	0.006126
26	841	0.000001	1079	0	1920	0.000841
27	980	0	1435	0.000021	2415	0.030135
28	970	0.000005	1560	0.000008	2530	0.01733
29	850	0.000002	1184	0.000003	2034	0.005252
30	1000	0	780	0	1780	0
31	1120	0.000007	1450	0.000009	2570	0.02089
Total :	30184		41485		71669	2.224619
Average :	974		1338		2312	3.1040E-05
Volume UF6 :	0.022	ft^3				

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5.4 Calculation of the Specific Activities

The "effective" specific activity of uranium is calculated using the expression in Eq. 1-2 and depends upon the enrichment of ^{235}U . Assuming an enrichment of 35% ^{235}U , the "effective" specific activity follows as

$$S = (0.4 + 0.38(35) + 0.0034(35)^2) \times 10^{-6} = 1.7865 \times 10^{-5} \text{ Ci/g.}$$

For December 1945, the "effective" activity of uranium is calculated by multiplying this value by the mass of uranium calculated in Section 5.3 and given as

$$A = (6.334)(1.7865 \times 10^{-5}) = 1.132 \times 10^{-4} \text{ Ci U.}$$

The specific activities of isotopic ^{234}U , ^{235}U , and ^{238}U are calculated using Eq 1-4. The decay constant, λ , for each isotope is given by Eq 5.4-1 as

$$\lambda_i = \frac{\ln 2}{T_{1/2}} \quad (5.4-1)$$

where $T_{1/2}$ is the half-life of the radioisotope.

The decay constant is typically expressed in units of s^{-1} and therefore requires conversion of the half-life to units of s. For the ^{234}U isotope, the decay constant is computed as

$$\lambda = \frac{\ln 2}{(2.46 \times 10^5)(365.25)(24)(3600)} = 8.92866 \times 10^{-14} \text{ s}^{-1}.$$

The decay constants for ^{235}U and ^{238}U are similarly calculated. Using Eq 1-4 and converting units of Bq to units of Ci, the specific activity of isotopic ^{234}U is calculated as

$$S = \frac{(8.92866 \times 10^{-14})(6.022045 \times 10^{23})}{(234.040904)(3.7 \times 10^{10})} = 6.2092 \times 10^{-3} \text{ Ci/g.}$$

The specific activities of isotopic ^{235}U and ^{238}U are similarly calculated.

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J. J. ShonkaTitle Uranium Release Estimates for the ORGDP Purge Cascade 12/45-12/46 (signed original on file)5.5 Calculation of the Isotopic Activity and Mass of ^{234}U , ^{235}U , and ^{238}U

An estimate of the contribution to the total "effective" activity of each uranium isotopic constituent is calculated according to Eq 5.5-1.

$$A_i = f_i S \quad (5.5-1)$$

where f_i is the fraction of total activity for each isotopic constituent from Fig 1-2, and S is the specific activity from Eq 1-2.

The fraction of total "effective" activity, f_i , for each uranium isotope is read from the graph in Fig 1-2 at the assumed 35% ^{235}U enrichment. The accompanying table at the right details the fractions used for each isotope in this calculation. For December 1945, the contribution to the total "effective" activity by ^{234}U is

i	f_i
^{238}U	0.012
^{235}U	0.43
^{234}U	0.945

$$A = (0.945)(1.132 \times 10^{-4}) = 1.069 \times 10^{-4} \text{ Ci } ^{234}\text{U}.$$

The contributions by ^{235}U and ^{238}U are calculated similarly. The estimate of the mass of each uranium isotopic constituent is calculated using equation Eq 1-3 and uses the theoretical specific activities for each uranium isotope computed in Section 5.4. For December 1945, the mass of ^{234}U is calculated as

$$m = \frac{1.069 \times 10^{-4}}{6.2092 \times 10^{-3}} = 1.722 \times 10^{-2} \text{ g } ^{234}\text{U}.$$

The mass of ^{235}U and ^{238}U are calculated similarly. The masses of the three isotopes were summed and compared with the total uranium mass calculated in Section 5.3 to ensure close agreement. The ratio of the ^{235}U mass to the total U mass was computed to ensure an approximate 35% enrichment level. The activity fractions in the table above represent the values resulting from several iterative refinements. The final fraction values from Fig 1-2 used in this calculation result in an understatement of the total uranium mass by 0.36% of the estimate resulting from the van der Waals real gas equation. The uncertainties in the experimental data and the fitted equation in Fig 1-1 and in selecting values from the graph in Fig 1-2 contribute to the differences in the mass calculations. Also notable is the omission of the ^{234}U contribution to the U and UF_6 molecular weight calculations in Section 5.1 which impact the van der Waals real gas equation calculations in Section 5.3, the specific activity calculations in Section 5.4, and the mass calculations in Section 5.5.

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5.6 Discussion of the Results of the Calculation

This calculation estimates that during the thirteen month period between December 1945 and December 1946, about 51.28 g of moderately enriched uranium was vented to the atmosphere by the purge cascade. The monthly median during this thirteen month period was about 0.32 g uranium with a standard deviation of about 5.5 g uranium. This release occurred daily and constituted a total of about 916 μCi with a monthly median of about 5.6 μCi and a standard deviation of about 98.8 μCi .

Gaseous diffusion causes a greater percent increase in ^{234}U than ^{235}U due to the better separation factor for ^{234}U . The half-life of ^{234}U is four and five orders of magnitude shorter than ^{235}U and ^{238}U and thus constitutes a higher percent of the overall activity. At 35% ^{235}U enrichment, about 94.5% of the total activity is due to the presence of $^{234}\text{UF}_6$ even though the weight percent of ^{234}U at this enrichment is only about 0.27%.

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6. REFERENCES

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Date 11/8/95

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J. J. Shonka

Title Uranium Release Estimates for the ORGDP Purge Cascade 12/45-12/46 (signed original on file)

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12/1/95
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7. ELECTRONIC FILES

The following files are included on the diskette that accompanies this calculation.

<u>File Name</u>	<u>Description</u>
95010R0.DOC	This document (Microsoft® Word version 6.0a).
95010R0.XLS	Spreadsheet for the calculation of the ORGDP purge cascade uranium release estimates for December 1945 through December 1946 (Microsoft® Excel version 5.0a).
RAWPURGE.XLS	Spreadsheet for the raw purge rate data from the K-25 Process Division Daily Reports (Microsoft® Excel version 5.0a).

Attachment 1

**Purge Rates November 1945 (partial) - December 1946
(from the K-25 Process Division Daily Reports at Site Records)**

Day	Month <u>DECEMBER 1945</u>				Month <u>NOVEMBER 1945</u>			
	Cell <u>312-2</u>		Cell <u>312-3</u>		Cell <u>312-2</u>		Cell <u>312-3</u>	
	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	Tops Conc
1	710	32e-6	1300					
2	470	40e-6	1900	—				
3	470	28e-6	1400	—				
4	1100	30e-6	1800	nil				
5	1210	7e-6	2360	56e-6				
6	1187	24e-6	3065	50e-6				
7	1020	10e-6	1395	12e-6				
8	840	10e-6	1450	nil				
9	1125	20e-6	1435	20e-6				
10	860	136e-6	1065	nil				
11	1160	50e-6	720	10e-6				
12	680	30e-6	980	25e-6				
13	385	32e-6	690	40e-6				
14	1100	11e-6	900	9e-6				
15	1040	100e-6	1020	80e-6				
16	1360	6e-6	540	1420e-6				
17	1160	15e-6	1300	4e-6				
18	1500	40e-6	990	50e-6				
19	960	2e-6	840	nil				
20	970	3e-6	610	56e-6				
21	820	3e-6	1220	35e-6				
22	1034	30e-6	2052	31e-6	1164	—	900	—
23	947	6e-6	2465	30e-6	1226	—	960	—
24	1108	6e-6	1455	3e-6	1125	—	700	—
25	997	3e-6	1045	3e-6	1100	—	900	—
26	841	1e-6	1079	nil	750	—	950	—
27	980	nil	1435	21e-6	900	—	1000	—
28	970	5e-6	1560	8e-6	1000	—	1250	—
29	850	2e-6	1134	3e-6	1100	—	1800	—
30	1000	nil	780	nil	600	88e-6	1600	nit
31	1120	7e-6	1450	9e-6	600	88e-6	1600	nit

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Day	Month <u>FEB 1946</u>				Month <u>MAY 1946</u>			
	Cell		Cell		Cell		Cell	
	<u>312-1 312-1</u>		<u>312-1 312-2</u>		<u>312-1</u>		<u>312-3</u>	
	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	Tops Conc
1	1570	6e-6	335*	1e-6*	1220*	12e-6*	1420	24e-6
2	1450	16e-6	115*	1e-6*	1200*	1e-6*	1000	36e-6
3	1000	12e-6	530*	73e-6*	1250*	1e-6*	1440*	1e-6
4	1110	1e-6	670*	4e-6*	880*	2e-6*	1440	20e-6
5	1135	20e-6	520*	8e-6*	880*	3e-6*	1440	120e-6
6	1200	2e-6	1000*	10e-6*	880*	43e-6*	1170	10e-6
7	1425	1e-6	750*	1e-6*	950*	244e-6*	900	55e-6
8	1190	1e-6	1110*	20e-6*	1440*	2e-6*	1080	30e-6
9	1200	22e-6	1600*	200e-6*	1420*	30e-6*	855	1e-6
10	2825	2e-6	5450*	10e-6*	4025*	125e-6*	2875	1e-6
11	1925	6e-6	4400*	20e-6*	2310	4e-6*	2940	4e-6
12	2160	2e-6	2625*	2e-6*	1200*	1e-6*	1260	10e-6
13	1810	1e-6	2050*	25e-6*	1460*	1e-6*	495	40e-6
14	2040	25e-6	2500*	4e-6*	1310*	1e-6*	555	1e-6
15	1560	31e-6	1600*	1e-6*	1340*	3e-6*	580	3e-6
16	1500	82e-6	1150*	1e-6*	1310*	1e-6*	1080	4e-6
17	1530	33e-6	994*	1e-6*	1360*	424e-6*	1390	100e-6
18	1648	54e-6	488*	2e-6*	1340*	25e-6*	1600	800e-6
19	1620	49e-6	1610*	1e-6*	1218*	1e-6*	682	1e-6
20	1310	27e-6	850*	2e-6*	1120*	4e-6*	1820	1e-6
21	1220	1e-6	574*	1e-6*	1080	5e-6*	140	1e-6
22	1230	3e-6	1200*	2e-6*	1265	90e-6	365	1e-6
23	1620	23e-6	1400*	2e-6*	1240	2150e-6	760	1e-6
24	1240	7e-6	0*	0*	1080	70e-6	575	50e-6
25	1280	110e-6	1350*	1e-6*	1600	30e-6	1250	1e-6
26	1540	25e-6	3000	2e-6	1080	36e-6	330	1e-6
27	1390	15e-6	860	1e-6	1290	31e-6	410	7e-6
28	1190	5e-6	1070	2e-6	945	510e-6	1300	3e-6
29					1055	3e-6	620	24e-6
30					1025	3e-6	41000	1e-6
31					1800	3e-6	400	1e-6

+1533 sf @ k-27
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 +1450 sf @ k-27
 +1105 @ k-27
 sf

+ 440 sf @ 312-

* k-312-3

* k-312-2

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Day	Month <u>APRIL 1946</u>				Month <u>MARCH 1946</u>			
	Cell <u>312-1</u>		Cell <u>312-2</u>		Cell <u>312-1</u>		Cell <u>312-2</u>	
	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	Tops Conc
1	2385	1e-6	2425	—	1290	19e-6	7270	21e-6
2	1650	1e-6	2700	—	1140	42e-6	1635	20e-6
3	1530	1e-6	2780	1e-6	1085	10e-6	1200	5e-6
4	1540	1e-6	2500	2e-6	1180	7e-6	1220	8e-6
5	1430	nil	2370	8e-6	1225	8e-6	2250	60e-6
6	1605	1e-6	2350	1e-6	1150	8e-6	875	95e-6
7	1340	1e-6	2055	1e-6	1180	8e-6	800	10e-6
8	1460	2e-6	1500	3e-6	1220	37e-6	685	30e-6
9	1380	11e-6	1880	1e-6	1250	2e-6	1960	30e-6
10	1590	1e-6	2790		1130	1e-6	1510	20e-6
11	1266		1319		1100	1e-6	425	35e-6
12	1300		1400		1080	70e-6	490	100e-6
13	1190		1350		1100	3e-6	1050	70e-6
14	1390		2900		1330	1e-6	2550	4e-6
15	1430		3000		1375		3250	40e-6
16	1450		1330	↓	1410		2900	1e-6
17	1230		1340	100e-6	1320		1830	↓
18	1200		1340	3e-6	1360	↓	3260	↓
19	1176		1340	1e-6	1310	1e-6	1920	1e-6
20	1210		1770	3e-6	1300	40e-6	2750	5e-6
21	1220		2330	1e-6	4300	1e-6	4360	100e-6
22	1100		1690		1940	1e-6	3415	20e-6
23	1285	↓	2325		1800	8e-6	3600	12e-6
24	1375	3e-6	2060		2050	9e-6	4100	9e-6
25	1280	1e-6	1620		1630	1e-6	4325	4e-6
26	1380		1580		1590	3e-6	3525	60e-6
27	1164		1350	↓	1540	1e-6	2925	1e-6
28	1090		1180	100e-6	1450	1e-6	2540	1e-6
29	1225		1310	1e-6	1460	7e-6	2850	11e-6
30	1160	100e-6	1310	1e-6	1610	1e-6	3000	5e-6
31					1450	1e-6	2850	1e-6

+ 750 scf
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Day	Month <u>JUNE 1946</u>				Month <u>MAY 1946</u>			
	Cell <u>312-1</u>		Cell <u>312-2</u>		Cell <u>312-1</u>		Cell <u>312-2</u>	
	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	Tops Conc
1	1180	1e-6	1330	1e-6	1160	5e-6	1500	1e-6
2	1200		1490		1330	1e-6	1630	1e-6
3	1250		1140		1240	1e-6	1260	1e-6
4	1360		960		1140	1e-6	1460	1e-6
5	1300		1100		1200	1e-6	1530	1e-6
6	1420		1150		1180	1e-6	1320	1e-6
7	1160	↓	1280	↓	1230	1e-6	1220	nil
8	1090	nil	600	nil	1450	1e-6	1370	1e-6
9	1610	nil	584	nil	1890	1e-6	4300	400e-6
10	1120	9e-6	1100	nil	1390	1e-6	2800	1e-6
11	1050	1e-6	840	1e-6	1600	nil	3420	nil
12	1020		1170		1530	nil	1260	8e-6
13	1030		800		1540	1e-6	1590	8e-6
14	1060		1720		1430	1e-6	2550	4e-6
15	1050		1390		1340	1e-6	2370	1e-6
16	1020		730		1200	1e-6	1540	1e-6
17	980		665		1420	1e-6	1250	1e-6
18	930		1430		1565	1e-6	1300	1e-6
19	1090		2260		1690	1e-6	1175	1e-6
20	1110		2080	↓	1635	1e-6	1375	1e-6
21	1130		2070	2e-6	1360	1e-6	2030	nil
22	1080		1290	1e-6	1170	nil	1190	nil
23	900		1210		1200	nil	1570	nil
24	910		1460		1390	nil	3400	nil
25	910		1510		1500	1e-6	4520	1e-6
26	1180		2000		1220		1800	↓
27	1080		1270		1256		1200	↓
28	1090		1060		1370		1135	14e-6
29	1080		1220		1560		890	1e-6
30	1060	↓	1810	↓	1220		1090	1e-6
31					1190	↓	1130	2e-6

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Day	Month <u>JULY 1946</u>				Month <u>AUGUST 1946</u>			
	Cell <u>312-1</u>		Cell <u>312-2</u>		Cell <u>312-1</u>		Cell <u>312-2</u>	
	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	Tops Conc
1	1180	1e-6	2100	1e-6	880	1e-6	2080	1e-6
2	1080	"	1440	"	857		830	
3	910	"	1260		1045		3237	
4	850	2e-6	1460		950		2654	
5	1000	1e-6	1090		806		2010	
6	1450		1200		800		1670	
7	1000		1560		1090		2960	
8	940		1680		1440		3400	
9	750		1380		841		2308	
10	850		1430		850		1690	
11	1070		2210		250		1300	
12	860		1150		760		1290	
13	1300		1810		880		1440	
14	860		1100		900		2090	
15	1020		1830		740		1230	
16	920		1930		790		1160	
17	910		1470		879		1220	
18	1010		1380		821		1390	
19	750		1710	✓	788		1885	
20	950		2660	4e-6	790		2500	
21	930		2390	1e-6	1140		1700	
22	820		2050		930		1290	
23	850		1630		850		1390	
24	980		2250		770		1240	
25	920		3380		730		1300	
26	1010		2560		880		1850	
27	1020		2880		1330		2680	
28	950		1980		920		2090	
29	1000		2800		1830		2270	
30	1070		2560		1210		2360	
31	930	✓	2260	✓	880	✓	1520	✓

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Day	Month <u>SEPT 1946</u>				Month <u>OCT 1946</u>			
	Cell <u>312-1</u>		Cell <u>312-2</u>		Cell <u>312-1</u>		Cell <u>312-3</u>	
	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	Tops Conc
1	865	1e-6	1500	1e-6	593	1e-6	1180	1e-6
2	850		1480		674		1289	
3	750		1640		649		1399	
4	400*	*	1780		690		1040	
5	400*	*	2400		680		1335	
6	1640		2300		683		1220	
7	1000		2370		751		1621	
8	1000		2910		654		1172	
9	1000		1840		654		2011	
10	800		1500		396		2575	
11	600		1970		483		1920	
12	1150		3600		397		1682	
13	1000		1500		397		1494	
14	590		1910*	*	341		1463	
15	680		1840*	*	530		2682	
16	590		1680*	*	297		2336	
17	750		2500*	*	631**	**	4754	
18	1020		3540*	*	691**	**	3820	
19	960		3140*	*	509**	**	1668	
20	720		1820*	*	495		2598	
21	690		1800*	*	340		616**	**
22	770		1980*	*	425		4856**	**
23	965		3180*	*	391		5350**	**
24	820		2140*	*	411		2387**	**
25	1200		2500*	*	311		1762**	**
26	850		1990*	*	671		4442**	**
27	1018		2083*	*	345		3910**	**
28	820		1895*	*	265		2165**	**
29	638		1809*	*	358		2783**	**
30	814		2266*	*	218		2926**	**
31					221		6080**	**

+1500 scf @ 312-3

+450 scf @ 312-2

* K-312-3

** K-312-2

*

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Day	Month <u>NOV 1946</u>				Month <u>DEC 1946</u>			
	Cell <u>312-1</u>		Cell <u>312-2</u>		Cell <u>312-1</u>		Cell <u>312-2</u>	
	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	Tops Conc
1	161	1e-6	2165	1e-6	—	—	2931	1e-6
2	121		3126		—	—	2931	1e-6
3	74		1767		—	—	3167	
4	106		2390		—	—	3138	
5	368	↓	2315		—	—	3121	
6	—	—	2402		—	—	3058	
7	—	—	2909		—	—	3075	
8	—	—	2980		—	—	3893	
9	—	—	2905		—	—	3479	
10	—	—	2918		—	—	3039	
11	—	—	2803		—	—	3149	
12	—	—	2985		—	—	3087	
13	—	—	3120		—	—	3678	
14	—	—	3997	↑	—	—	3476	
15	—	—	4487		—	—	2943	
16	—	—	3173		—	—	3905	
17	—	—	3002		—	—	4222	
18	—	—	3168		—	—	5422	
19	—	—	3534		—	—	5124	
20	—	—	3102		—	—	5020	
21	—	—	3977		—	—	3851	
22	—	—	4680		—	—	2983	
23	—	—	3225		—	—	2810	
24	—	—	3081		—	—	4059	
25	—	—	3041		—	—	4713	
26	—	—	3310		—	—	3270	
27	—	—	3367		—	—	4652	
28	—	—	3156		—	—	2520	
29	—	—	3006		—	—	4067	
30	—	—	3104		—	—	2964	
31				↓	—	—	2924	↓

312-2 Represents Total Purge

OST #3189

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Calculation Control Sheet

Calculation number: SPA-95-011 REV. 0

Title: URANIUM RELEASE ESTIMATES FOR THE ORGDP PURGE
CASCADE 1961

Reason for calculation/revision: ORIGINAL

Client: HEMLOCK, INC.

Project: OAKRIDGE HEALTH STUDIES

Project/Task Number: TASK 6

Prepared by: TE BENNETT

Date: 11/10/95

Independent Technical Review by:

[Signature]

Date: 12/1/95

Quality Assurance Review by:

Deborah B. Shonka

Date: 12/4/95

☐ This calculation has been voided or superseded by _____
(calculation number)

This document has been reviewed for classification and has been determined to be UNCLASSIFIED.
<u>Thomas W. Selby</u> ADC Signature
<u>12/7/95</u> Date

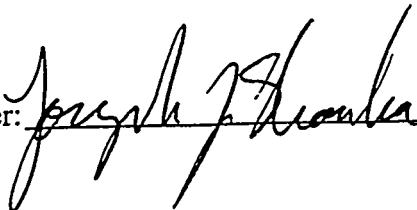
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Review Method Sheet

The undersigned has reviewed this calculation in accordance with the method(s) indicated below.

1. Computer Aided Calculation	
a	Review to determine that the computer program(s) has been validated and documented, is suitable to the problem being analyzed, and that the calculation contains all necessary information for reconstruction at a later date.
b	Review to determine that the input data as specified for program execution is consistent with the design input, correctly defines the problem for the computer algorithm and is sufficiently accurate to produce results within any numerical limitations of the program.
c	Review to verify that the results obtained from the program are correct and within stated assumptions and limitations of the program and are consistent with the input.
d	Review validation documentation for temporary changes to listed, or developmental, or unique single application programs, to assure that the methods used adequately validate the program for the intended application.
e	Review of code input only, since the computer program has sufficient history of use at Shonka Research Associates, Inc. in similar calculations.
f	Review arithmetic necessary to prepare code input data.
g	Other:
2. Hand Prepared Calculations	
<input checked="" type="checkbox"/> a	Detailed review of the original calculations.
<input checked="" type="checkbox"/> b	Review by an alternate, simplified, or approximate method of calculation.
c	Review of a representative sample of repetitive calculations.
d	Review of the calculation against a similar calculation previously performed.
e	Other:
3. Revisions	
a	Editorial changes only
b	Elimination of unapproved input data without altering calculated results.
c	Other:
4. Other	

Reviewer:



Date:

12/1/85

CALC NO SRA-95-011 REV 0Project/Task ChemRisk/TDH Oak Ridge Dose ReconstructionPrepared by T. E. BennettDate 11/8/95

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12/1/95
J. J. ShonkaTitle Uranium Release Estimates for the ORGDP Purge Cascade 1961gjs
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ABSTRACT

This calculation represents the third in a series of calculations aimed at establishing a methodology and strategy in analyzing the quality of the historical effluent monitoring data and practices at the Oak Ridge Gaseous Diffusion Plant. Within that scope, the emission rates from the purge cascade during the months of March 1961 through December 1961 were analyzed in order to determine its magnitude as a site contributor in the total historical uranium emissions. Daily purge rate data that documented the volumetric flow of the purge gas and its concentration of UF_6 were used to compute the daily flow of UF_6 in the purge cascade. The total volumetric flow of UF_6 for each month was used in the modified van der Waals real gas equation in order to estimate the mass of UF_6 released. Calculations show that over the ten month period, nearly 2.3 kg of highly enriched UF_6 was measured by the space recorder monitoring instrumentation located in cells K-312-1, K-312-2, K-311-1 of the purge cascade. This corresponds to the release of about 100 mCi of alpha activity and includes contributions from the mixture of $^{234}\text{UF}_6$, $^{235}\text{UF}_6$, and $^{238}\text{UF}_6$. These estimates do not include any reductions due to carbon or alumina traps located downstream of the space recorder instrumentation prior to the purge stack exit.

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Attachments

1. Purge Rates February 1961 (partial) - December 1961 (from the K-25 Area 5 Foreman Logbooks at K-1034-A Site Records)

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1. INTRODUCTION

Task 6 of the Oak Ridge Dose Reconstruction focuses on the evaluation of the quality of historical airborne and waterborne effluent monitoring data and the determination of the potential significance of unmonitored emissions. Uranium played an important role throughout historical operations on the Oak Ridge Reservation (ORR) and is known to have been released to the environment through air and water. The two largest uses of uranium on the Reservation were the enrichment processes of the ^{235}U isotope by electromagnetic separation at the Y-12 facility and gaseous diffusion at the K-25 facility.

Task 6 activities will be directed at establishing revised uranium release estimates with an associated uncertainty over that of the screening analyses conducted during the Dose Reconstruction Feasibility Study. These activities will support refined assessment of the potential magnitude of health hazards from historical uranium exposures based on both the chemical and radiotoxicity of uranium.

This calculation represents the third in a series of calculations aimed at establishing a methodology and strategy in analyzing the quality of the historical effluent monitoring data and practices at the Oak Ridge Gaseous Diffusion Plant (ORGDP). For many years the purge cascade represented the only on-site monitored emission source. This along with previous and subsequent calculations will provide a historical review of the uranium released from the ORGDP purge cascade including an assessment of the potential uncertainties and biases in the measurement and computation of the estimates.

Light molecular weight gases were purged from the top of the diffusion cascade that would otherwise block the withdrawal of enriched UF_6 product. These light gases originated from the following sources.

- *nitrogen* - mainly from the inleakage occurring at every pump shaft seal in the diffusion cascade
- *hydrogen fluoride* - from reaction of inleaking moist air with UF_6
- *oxygen, argon* - from inleaking air
- *chlorine fluorides* - used in conditioning and drying of metal surfaces
- *fluorine* - used in conditioning of metal surfaces
- *coolant vapor* - inleakage from the compressor and pump coolant system

The light gases in the process stream had a molecular weight substantially less than that of the UF_6 component and were carried along effectively by the diffusion process to the top of the cascade. A section at the top of the cascade just above the product withdrawal point was reserved as the purge cascade. The purge cascade separated these light gases from the enriched UF_6 product and vented them to the atmosphere. The purge cascade usually existed in two sections: a side purge and a top

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purge. The side purge separated the intermediate molecular weight gases (*i.e.* coolant vapor, chlorine fluorides, etc.) from the UF_6 . The top purge separated the remaining lighter gases. At various times throughout the operational history, the side purge was routed to the top purge. The effluents were pumped through traps in order to reduce the uranium content before venting to the atmosphere. In the earlier years these traps consisted only of carbon and alumina, but later sodium fluoride (NaF) and potassium hydroxide (KOH) scrubbers were added. Because of the large difference in molecular weights between the light gases and UF_6 , only a few diffusion stages were needed to effectively perform the separation. Similar to the main diffusion cascade, the purge cascade consisted of compressors, converters, motors, coolers, piping, control and block valves, and instrumentation. The major difference between the main diffusion cascade and the purge cascade was the smaller amount of UF_6 flow (MMES 1985).

The problem of analyzing the gas in the purge cascade was complicated by the fact that the UF_6 concentration varied greatly from one end of the cascade to the other. Near the bottom of the cascade the process stream consisted of essentially pure UF_6 , whereas at the cascade top the stream consisted of light gases containing only small traces of UF_6 . The line recorder was designed for analyzing UF_6 containing relatively small amounts of impurities. The method it employed measured the flow of gas to a mass spectrometer tube by means of a Pirani gauge flowmeter; the UF_6 was removed chemically before reaching the spectrometer tube and the residual gas concentration measured by means of the spectrometer tube. In analyzing for UF_6 in the presence of relatively large amounts of impurities, the accuracy dropped sharply. Due to the corrosive nature of UF_6 , the concentration could not be directly measured by the spectrometer tube. Therefore, the UF_6 concentration was determined by the difference between the flow computed from the flowmeter and the remaining light gas concentration determined by the mass spectrometer tube reading. Only with careful calibration was it possible to determine either of these quantities with an accuracy of 1% or better. Accordingly, the line recorder became practically useless for determining the composition of a mixture containing under 2% of UF_6 (OM-48 1945).

In order to supplement the line recorder in the purge cascade, an instrument known as the space recorder was developed. The principal component of the space recorder was an ionization chamber more commonly referred to as the "signal can." The signal can measures the specific radioactivity of the gas present, and since UF_6 is an alpha emitter, this method provided a convenient means for measuring the UF_6 content of gas samples. The space recorder could detect the presence of mol fractions of UF_6 in the light gas purge of the order of 10^{-6} (OM-48 1945). The radioactivity of UF_6 consists of the emission of high energy alpha particles at a definite uniform rate. This rate depends upon the relative isotopic composition, since all three isotopes emit alpha particles at a different rate. These alpha particles as emitted have a definite range of travel, which is inversely proportional to the pressure. In the gas samples, at standard temperature and pressure, this range of travel is on average approximately 3 cm. While travelling this distance the energy of the emitted particle is expended by the production of about 130,000 ions resulting from collision of the particles with gas molecules.

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The collection of these ions and a measurement of the resultant electrical current constitutes a determination of the number of particles present and hence a determination of the UF_6 concentration. A record of the purge gas volumetric flow and the concentration of UF_6 in the purge stream during the 1961 time frame was reported in the Area 5 Foreman Logbook.

This calculation analyzes a ten month span of daily purge cascade effluent data for the months of March 1961 through December 1961. These estimates do not represent an actual estimation of the amount of uranium released to the atmosphere during this time period. The sample withdrawal point for the space recorders were located upstream of the carbon and alumina traps. The traps would have removed much of the uranium prior to the venting of the purge gases to the atmosphere and these benefits are not included in these estimates. With this limitation in mind, this calculation makes liberal use of the terms "release", "released", "vented", and "effluent." Furthermore, these estimations are also subject to certain assumptions, biases, and other uncertainties. Several assumptions and potential sources of uncertainty will be presented in this calculation, but the analyses to quantify the impact of these assumptions to the results of this calculation will be contained in subsequent calculations.

In this calculation, data sheets containing the daily purge rates for the months of March 1961 through December 1961 were transcribed from the logbooks maintained by the Area 5 Foreman and transferred to spreadsheets (Purge Rates 1961). The volume of gas purged each day and its UF_6 concentration was used to compute the daily volumetric flow of UF_6 released. The daily flow of UF_6 was summed to compute the estimate of the total volume of UF_6 vented during the month. The mass of UF_6 released each month in the purge cascade was derived from this known volume at standard conditions by application of the modified van der Waals real gas equation (Ackley, Magnuson 1951) as given in Eq. 1-1,

$$m = \frac{P(1 + AP)V}{RT} \quad (1-1)$$

where P is the pressure of the gas,

A is the temperature-dependent van der Waals coefficient for UF_6 ,

V is the volume of the gas,

R is the UF_6 gas constant, and

T is the temperature of the gas.

The activity of UF_6 released each month in the purge cascade was computed by multiplying the grams of UF_6 by the specific activity of UF_6 at the assumed ^{235}U enrichment level. The "effective" specific activity of a mixture of $^{234}\text{UF}_6$, $^{235}\text{UF}_6$, and $^{238}\text{UF}_6$ (as found in the purge cascade effluent) follows Eq. 1-2.

$$S = (0.4 + 0.38E + 0.0034E^2) \times 10^{-6} \text{ Ci/g} \quad (1-2)$$

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where E is the percent ^{235}U by weight. Eq. 1-2 is fitted to the experimental data in Fig 1-1 (Rich 1988). The contribution to the total "effective" activity of each isotope of uranium was determined from the graph presented in Fig 1-2 (Rich 1988) and used to determine the activity of each isotope. The mass of ^{234}U , ^{235}U , and ^{238}U in the purge effluent could then be calculated from its activity and theoretical specific activity as given by Eq. 1-3. The results of the isotopic mass calculations were compared to the mass calculations for UF_6 using Eq. 1-1 in order to determine the "goodness" of the values selected from Fig 1-2.

$$m_i = \frac{A_i}{S_i} \quad (1-3)$$

where A_i is the activity of the radioisotope, and
 S_i is the specific activity of the radioisotope.

The theoretical specific activity of each uranium isotope is calculated by Eq. 1-4.

$$S_i = \frac{\lambda_i N_A}{M_i} \quad (1-4)$$

where λ_i is the decay constant of the radioisotope,
 N_A is Avogadro's Number, and
 M_i is the atomic weight of the radioisotope.

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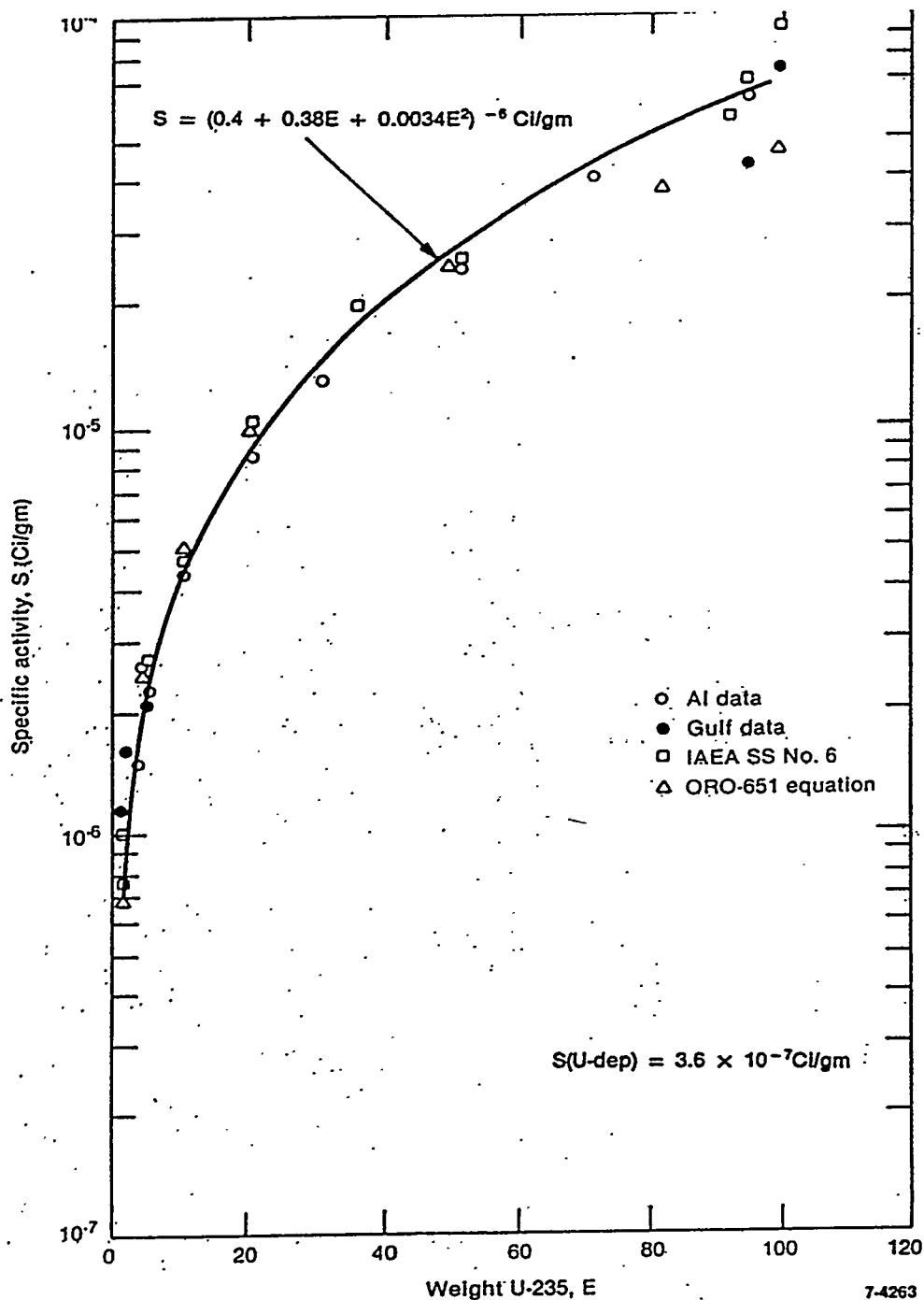


Fig 1- 1 : Specific Activity for Mixtures of ^{234}U , ^{235}U , and ^{238}U

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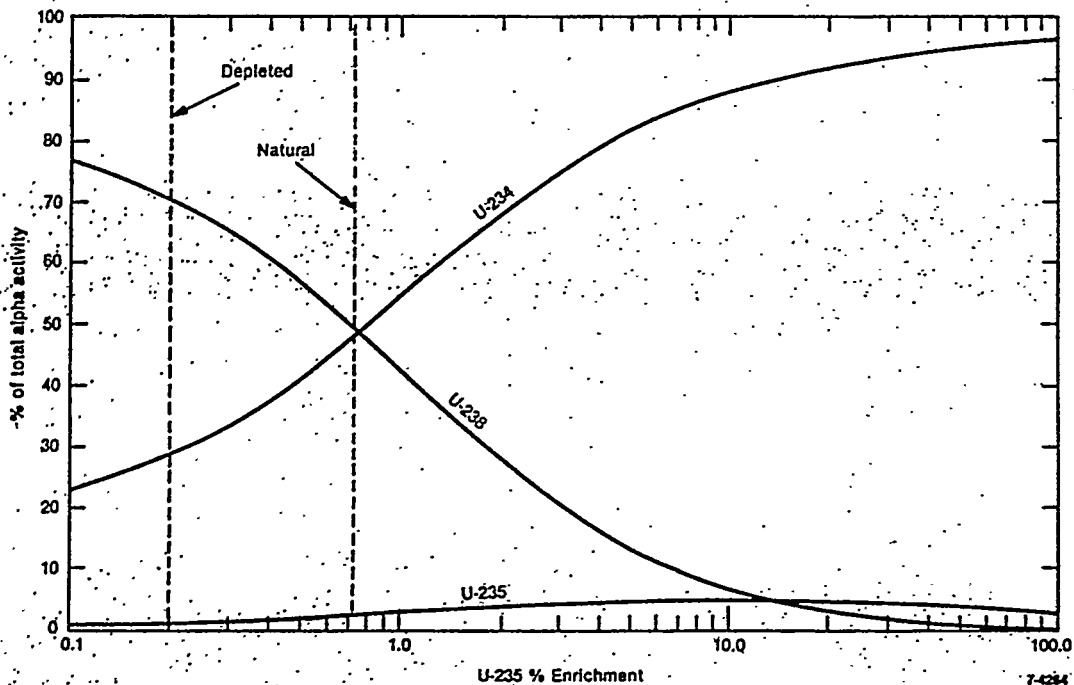


Fig 1-2 : Percent of total radioactivity by isotope vs. % weight ²³⁵U enrichment

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2. SUMMARY OF RESULTS

The monthly uranium release estimates from the ORGDP purge unit between March 1961 and December 1961 are presented below in Table 2-1. Totals are given for sum of all months. In addition, the median and standard deviation are given for all computations. The first column of data presents the results of the UF_6 mass calculations using the modified van der Waals real gas equation in Eq. 1-1. The mass of the F_6 is subtracted from the UF_6 mass and the results presented in the second column along with the computation of the "effective" activity given by Eq. 1-2. The next three columns of data present the mass and activity of ^{238}U , ^{235}U , and ^{234}U respectively. The activity of each of the three constituents are computed by applying the appropriate activity fraction from Fig 1-2 to the "effective" activity. The mass of each of the three constituents are computed using the theoretical specific activities of each radioisotope. The last column presents the sum of the masses of each of the three constituents and the percent difference with the uranium mass based upon the modified van der Waals real gas equation computation.

The total UF_6 release over the ten month period is 2.293 kg with 1.545 kg being uranium. This corresponds to a total "effective" activity of 100.6 mCi and assumes a 93% ^{235}U enrichment. The "effective" activity corresponds to alpha decay contributions from the mixture of ^{234}U , ^{235}U , and ^{238}U . According to Fig 1-2 at 93% ^{235}U enrichment, virtually all of the activity is due to the presence of ^{234}U . At this assumed enrichment, about 96.9% of the activity results from the ^{234}U isotope, 3.1% from the ^{235}U isotope, and the remaining from the ^{238}U isotope. The results also indicate that at this assumed ^{235}U enrichment, the ^{234}U isotope is enriched from 0.0056% natural abundance to just over 1% and the ^{238}U isotope is depleted to under 7%. The sum of the three isotopic masses overstate the calculated uranium mass from the van der Waals gas equation by 1.28%. This difference is subject to uncertainty in the experimental data and the fitted equation in Fig 1-1 and to uncertainty in reading the data from the graph in Fig 1-2.

These estimates do not include any reductions due to carbon or alumina traps located downstream of the space recorder instrumentation prior to the purge stack exit.

Table 2-1: Purge Cascade Uranium Release Estimates Mar 1961 - Dec 1961

	UF6	U		U-238		U-235		U-234		Total	
Month	[g]	[g]	[Ci]	[g]	[Ci]	[g]	[Ci]	[g]	[Ci]	[g]	Δ%
Mar-61	7.797E+01	5.252E+01	3.422E-03	3.555E+00	1.198E-06	4.910E+01	1.051E-04	5.338E-01	3.314E-03	5.320E+01	1.28%
Apr-61	8.140E+01	5.483E+01	3.572E-03	3.722E+00	1.250E-06	5.126E+01	1.107E-04	5.573E-01	3.460E-03	5.554E+01	1.28%
May-61	1.330E+02	8.961E+01	5.838E-03	6.082E+00	2.043E-06	8.377E+01	1.810E-04	9.107E-01	5.655E-03	9.076E+01	1.28%
Jun-61	6.954E+01	4.684E+01	3.052E-03	3.179E+00	1.068E-06	4.379E+01	9.460E-05	4.761E-01	2.956E-03	4.744E+01	1.28%
Jul-61	1.339E+02	9.021E+01	5.877E-03	6.122E+00	2.057E-06	8.433E+01	1.822E-04	9.168E-01	5.693E-03	9.136E+01	1.28%
Aug-61	4.434E+02	2.987E+02	1.946E-02	2.027E+01	6.811E-06	2.792E+02	6.033E-04	3.036E+00	1.885E-02	3.025E+02	1.28%
Sep-61	2.285E+02	1.539E+02	1.003E-02	1.044E+01	3.509E-06	1.439E+02	3.108E-04	1.564E+00	9.711E-03	1.559E+02	1.28%
Oct-61	5.132E+02	3.457E+02	2.252E-02	2.346E+01	7.882E-06	3.232E+02	6.981E-04	3.513E+00	2.181E-02	3.501E+02	1.28%
Nov-61	2.903E+02	1.956E+02	1.274E-02	1.327E+01	4.459E-06	1.828E+02	3.950E-04	1.988E+00	1.234E-02	1.981E+02	1.28%
Dec-61	3.222E+02	2.170E+02	1.414E-02	1.473E+01	4.948E-06	2.029E+02	4.383E-04	2.206E+00	1.369E-02	2.198E+02	1.28%
Total	2.293E+03	1.545E+03	1.006E-01	1.049E+02	3.523E-05	1.444E+03	3.120E-03	1.570E+01	9.749E-02	1.565E+03	1.28%
Median	1.812E+02	1.220E+02	7.951E-03	8.283E+00	2.783E-06	1.141E+02	2.465E-04	1.240E+00	7.702E-03		
Std Dev	1.508E+02	1.016E+02	6.619E-03	6.895E+00	2.316E-06	9.497E+01	2.052E-04	1.032E+00	6.411E-03		

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3. METHODS

The results summarized in Table 2-1 were based upon data sheets generated from the K-25 Area 5 Foreman Logbooks. These data sheets contain the log of the purge unit volumetric flow and UF_6 concentration for each of the three shifts as measured at purge cells K-312-1, K-312-2 and K-311-1, and are included in this calculation as Attachment 1. As part of this calculation, these data sheets were recreated as individual spreadsheets in a Microsoft[®] Excel workbook and are provided electronically in file 95011R0.XLS. In order to determine an estimate for the total amount of uranium released by the purge unit over this ten month period, a number of computations using the raw purge data were required.

During the 1961 time frame, the purge unit consisted of three cells, K-312-1, K-312-2, and K-311-1. At any one time, one or two of the cells were in operation and the remaining cell(s) in standby. One cell operated as a top purge and another cell operated as a side purge. Additionally, the side purge could be bypassed and routed to the top purge allowing the effective operation of the ORGDP using a single purge unit. The raw purge data for each day in the Area 5 Foreman Logbooks consisted of measurements during the night, day, and evening shifts. This three shift purge data must first be averaged to daily purge flows for each of the purge units. The purge gas flow from the one or two cells were summed to compute the total daily flow of purge gases. These daily purge gas flows were subsequently summed to compute the total purge gas flow for the month. This total was then divided by the number of days in the month to compute the average daily purge gas flow.

Likewise, the raw purge data for each day in the Area 5 Foreman Logbooks consisted of measurements of the UF_6 concentration in the purge gas during the night, day, and evening shifts. This three shift UF_6 concentration data must first be averaged to daily UF_6 concentrations for each of the purge units. The daily UF_6 volumetric flow was calculated by multiplying the UF_6 concentration and the purge gas volumetric flow. The results of this calculation for both the purge cells were summed to compute the total daily UF_6 volumetric flow in the purge unit. These daily UF_6 flows were summed to compute the total UF_6 volume released by the purge cascade for the month. This total was divided by the monthly total purge gas flow to compute the average daily UF_6 volumetric flow.

The mass of UF_6 released by the purge unit was calculated using Eq 1-1 and the monthly total volume of UF_6 , the modified van der Waals pressure, standard temperature, and the UF_6 gas constant. The UF_6 mass was multiplied by the ratio of the molecular weight of uranium to the molecular weight of UF_6 resulting in the computation of the uranium mass. The "effective" specific activity of uranium at the assumed 93% ^{235}U enrichment was calculated using Eq 1-2 and the result multiplied by the uranium mass to compute the "effective" activity of the uranium. The fractional contribution of this "effective" activity by ^{234}U , ^{235}U , and ^{238}U are obtained from the graph in Fig 1-2 and each multiplied by the "effective" activity to compute the estimated activity of each isotopic

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constituent. The theoretical specific activity of each constituent was calculated using Eq 1-4 and then divided into the respective activity estimates according to Eq 1-3 to compute the estimated mass of each isotopic constituent. The three masses of the constituents were summed and compared to the uranium mass based upon Eq 1-1 in order to ensure the activity fractions obtained from the graph in Fig 1-2 were appropriate. The activity fraction values were iteratively refined until good agreement between the two mass calculations were obtained.

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4. ASSUMPTIONS

- 4.1. The atomic weight of ^{238}U is given as 238.05077 g/mol (Physics 1967).
- 4.2. The half-life of ^{238}U is given as 4.46×10^9 yr (Physics 1967).
- 4.3. The atomic weight of ^{235}U is given as 235.043915 g/mol (Physics 1967).
- 4.4. The half-life of ^{235}U is given as 7.04×10^8 yr (Physics 1967).
- 4.5. The atomic weight of ^{234}U is given as 234.040904 g/mol (Physics 1967).
- 4.6. The half-life of ^{234}U is given as 2.46×10^5 yr (Physics 1967).
- 4.7. The atomic weight of fluorine is given as 18.998403 g/mol (Physics 1967).
- 4.8. Avogadro's Number is given as 6.022045×10^{23} atoms/mol (Physics 1967).
- 4.9. For purposes of U and UF_6 molecular weight calculations, the contribution due to $^{234}\text{UF}_6$ are assumed negligible.
- 4.10. Standard pressure is assumed 14.7 psia (Lee 1989).
- 4.11. Standard temperature is assumed 59 F or 519 R (Lee 1989).
- 4.12. The universal gas constant \bar{R} is given as $10.73 \text{ psia ft}^3 \text{ lb}^{-1} \text{ mol}^{-1} \text{ R}^{-1}$ (Black, Hartley 1985).
- 4.13. The uranium enrichment of the UF_6 in the purge stream for the purposes of this calculation is assumed to be 93% ^{235}U . During the time frame of this calculation, the ORGDP was engaged in process activities producing highly enriched uranium of ~93% ^{235}U . The precise enrichment characteristic of the product remains classified.
- 4.14. The conversion factor of 453.6 g/lb is used to convert between units of mass (Black, Hartley 1985).
- 4.15. The conversion factors of 365.25 days/yr, 24 hr/day, and 3600 s/hr are used to convert between units of time.
- 4.16. The conversion factor of 3.7×10^{10} Bq/Ci is used to convert between units of activity.
- 4.17. The UF_6 concentrations recorded on the purge rate data sheets are assumed given as mole weight percentages.

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- 4.18. Purge gas flow and UF_6 concentration measurements in the K-312-1, K-312-2 and K-311-1 cells represent the sum total of the flow in the purge unit.
- 4.19. This calculation assumes UF_6 in the purge stream behaves as a real gas following the behavior prescribed by the modified van der Waals gas equation. The van der Waals coefficient for UF_6 is a function of temperature with the values of 0.033 atm^{-1} at 141.7 F and 0.021 atm^{-1} at 201.0 F (Ackley, Magnuson 1951).
- 4.20. The trap efficiencies are assumed negligible, thus implying that the purge gas flow and UF_6 concentration recorded on the purge rate data sheets are identical to the material actually vented to the atmosphere. The impact to the uranium release estimates due to actual trap efficiency, sampling biases and losses, and measurement uncertainties will be addressed in subsequent calculations. Some of the relevant sampling issues are (1) the maintenance of sufficient sample line temperature to prevent UF_6 condensation, (2) losses in the sample lines due to the length and of any bends, and (3) measurement uncertainties in the space recorder.

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5. CALCULATION

5.1 Calculation of the UF₆ Gas Constant

The symbol R in Eq. 1-1 is called the *gas constant* and its value depends upon the particular gas being considered. The value of R for each gas is determined by the equation

$$R = \frac{\bar{R}}{M} \quad (5.1-1)$$

where \bar{R} is a physical constant called the *universal gas constant* and is given in Section 4. It is first necessary to compute the atomic weight of uranium. Eq 5.1-2 gives the molecular weight of an isotopic mixture as

$$\frac{1}{M} = \frac{1}{100} \sum \frac{w_i}{M_i}$$

and for 93% ²³⁵U

$$\frac{1}{M} = \frac{1}{100} \left(\frac{93}{235.043915} + \frac{7}{238.05077} \right),$$

which gives $M = 235.2519202$. The molecular weight of UF₆ is then $235.2519202 + (6)(18.998403) = 349.2423382$. Substituting into Eq. 5.1-1, the value of R for UF₆ becomes

$$R = \frac{10.73}{349.2423382} = 0.030723652 \text{ psia ft}^3 \text{ lb}^{-1} \text{ R}^{-1}.$$

5.2 Calculation of the Modified van der Waals Pressure

Since UF₆ behaves as a real gas, the modified van der Waals pressure is required to account for the non-ideality of the UF₆ in the gaseous diffusion process. The expression $P(1+AP)$ in Eq. 1-1 represents the modified the pressure in the traditional ideal gas equation. The parameter A in the above expression represents the temperature-dependent van der Waals coefficient. Given values for

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the van der Waals coefficient at two temperatures, the following expression was derived that describes the nature of the van der Waals coefficient (atm^{-1}) as a function of UF_6 temperature (F).

$$A = -2.02 \times 10^{-4} T + 0.0617 \text{ atm}^{-1}$$

Naturally, as the temperature of the gas increases, the gas behaves in a more ideal manner. For standard conditions in which the temperature is given as 59 F, the van der Waals coefficient is given as $0.049782 \text{ atm}^{-1}$, or 0.0034 psia^{-1} . Using the van der Waals coefficient at standard conditions, the above expression evaluates the modified van der Waals pressure as

$$P' = 14.7 \times (1 + (0.0034)(14.7)) = 14.75 \text{ psia.}$$

5.3 Calculation of the Monthly UF_6 and Uranium Release Estimate

Table 5.1-1 depicts a sample spreadsheet of the raw purge data for a three shift daily log from the Area 5 Foreman Logbook for the month of March 1961. The spreadsheets for the remaining months are in the Microsoft® Excel workbook RAWPURGE.XLS. The three data points for each day were averaged in order to provide daily averages of the purge rate (scfd) and Tops Conc (mol wt.% UF_6). For example, the daily average purge rate for March 1, 1961 would be

$$\frac{5909 + 6101 + 5968}{3} = 5993 \text{ scfd,}$$

and the daily average UF_6 concentration for March 1, 1961 would be

$$\frac{0.000034 + 0.0002 + 0.000065}{3} = 9.96667 \times 10^{-5} \text{ mol wt. \% } \text{UF}_6.$$

Table 5.1-2 depicts a sample daily purge rate data spreadsheet for the month of March 1961. The spreadsheets for the remaining months are in the Microsoft® Excel workbook 95011R0.XLS. The daily purge rates on the data sheets are given in units of standard cubic feet per day (scfd) and the Tops Conc are given units of mol wt % UF_6 .

For each day, the total purge gas volumetric flow and UF_6 concentration is calculated. The purge flow is the sum of the purge rate in the top (312-1) and side (312-2) purge units. The daily molecular weighted UF_6 fraction in the purge gases is computed by multiplying the purge rate and the tops concentration and summing this value in K-312-1 and K-312-2. These daily rates are shown in the rightmost column in Table 5.1-2. For example, the per cent volume of UF_6 purged on March 1, 1961 would be calculated as

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$$(5993 \times 9.9667 \times 10^{-5}) + (0 \times 0) = 0.59726911 \% \text{ scfd.}$$

Table 5.1-1: Raw Three Shift Purge Data Spreadsheet for March 1961

Cell 312-1						
12 am - 8 am			8 am - 4 pm		4 pm - 12 pm	
Day	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	Tops Conc
1	5909	0.000034	6101	0.0002	5968	0.000065
2	6101	0.000058	6174	0.000105	6189	0.000036
3	5983	0.000109	6086	0.000061	6145	0.000065
4	5954	0.000205	6159	0.000079	6101	0.000018
5	5807	0.000035	5924	0.000096	6101	0.000107
6	5821	0.00003	6042	0.000091	6189	0.000062
7	5865	0.00003	6114	0.000267	6115	0.000163
8	6012	0.0002	6262	0.000079	6189	0.000025
9	6027	0.00001	6145	0.000024	6100	0.00004
10	5998	0.000015	6012	0.000019	6189	0.000109
11	6233	0.000014	5880	0.00001	6071	0.000008
12	6203	0.000015	6145	0.000073	6056	0.000008
13	6071	0.000089	6086	0.000031	5880	0.000049
14	5630	0.000034	5557	0.000105	5557	0.000098
15	6321	0.000036	5967	0.000044	6174	0.000078
16	6248	0.00003	5792	0.000065	6703	0.000044
17	6600	0.000035	6424	0.00003	6468	0.000021
18	6733	0.000123	6777	0.00001	6615	0.000014
19	6630	0.000022	6615	0.00004	6644	0.000034
20	6557	0.000014	6115	0.00008	6262	0.00027
21	6380	0.000273	6380	0.000015	6409	0.00003
22	6703	0.000047	6777	0.000055	7027	0.000076
23	6983	0.000015	7159	0.000057	7409	0
24	6571	0.000005	6048	0.00001	5953	0.00001
25	6189	0.000025	6189	0.000053	6306	0.00006
26	6468	0.000016	6497	0.000048	6365	0.0001
27	6615	0.00011	6512	0.000078	6453	0.0001
28	6497	0.000051	6424	0.000024	6586	0.000175
29	6350	0.000038	6365	0.000066	6439	0.00037
30	6512	0.000028	6630	0.000062	6733	0.000076
31	6802	0.001224	6895	0.001223	6762	0.000205

The total volume of UF_6 purged for the month is the sum of these daily volumetric flows divided by 100. For March 1961, this computes to 0.186 ft^3 . The total volume of UF_6 at standard conditions is used in Eq. 1-1 to estimate the mass of UF_6 and uranium released from the purge unit for the month. The modified van der Waals pressure is substituted for the expression $P(1+AP)$. This calculation for March 1961 precedes as follows

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$$m = \frac{(14.75)(0.186)(453.6)}{(0.030723652)(519)} = 77.97 \text{ g UF}_6.$$

The uranium-only fraction of the UF_6 mass is calculated by multiplying by the ratio of the molecular weights of U and UF_6 computed in Section 5.1. For March 1961, the calculation precedes as

$$m = \frac{(77.97)(235.2519202)}{349.2423382} = 52.52 \text{ g U.}$$

5.4 Calculation of the Specific Activities

The "effective specific activity of uranium is calculated using the expression in Eq. 1-2 and depends upon the enrichment of ^{235}U . Assuming an enrichment of 93% ^{235}U , the specific activity follows as

$$S = (0.4 + 0.38(93) + 0.0034(93)^2) \times 10^{-6} = 6.5147 \times 10^{-5} \text{ Ci/g.}$$

For March 1961, the "effective" activity of uranium is calculated by multiplying this value by the mass of uranium calculated in Section 5.3 and given as

$$A = (52.52)(6.5147 \times 10^{-5}) = 3.422 \times 10^{-3} \text{ Ci U.}$$

The specific activities of isotopic ^{234}U , ^{235}U , and ^{238}U are calculated using Eq 1-4. The decay constant, λ_i , for each isotope is given by Eq 5.4-1 as

$$\lambda_i = \frac{\ln 2}{T_{1/2}} \quad (5.4-1)$$

where $T_{1/2}$ is the half-life of the radioisotope.

The decay constant is typically expressed in units of s^{-1} and therefore requires conversion of the half-life to units of s. For the ^{234}U isotope, the decay constant is computed as

$$\lambda = \frac{\ln 2}{(2.46 \times 10^5)(365.25)(24)(3600)} = 8.92866 \times 10^{-14} \text{ s}^{-1}.$$

The decay constants for ^{235}U and ^{238}U are similarly calculated. Using Eq 1-4 and converting units of Bq to units of Ci, the specific activity of isotopic ^{234}U is calculated as

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$$S = \frac{(8.92866 \times 10^{-14})(6.022045 \times 10^{23})}{(234.040904)(3.7 \times 10^{10})} = 6.2092 \times 10^{-3} \text{ Ci/g.}$$

The specific activities of isotopic ^{235}U and ^{238}U are similarly calculated.

Table 5.1-2: Spreadsheet of Purge Rate Data for March 1961

Day of Month	312-1		312-2		Total	Mol% UF6
	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	
1	5993	9.96667E-05	0	0	5993	0.597269111
2	6155	6.63333E-05	0	0	6155	0.408259556
3	6071	7.83333E-05	0	0	6071	0.475587778
4	6071	0.000100667	0	1.33333E-05	6071	0.611180889
5	5944	7.93333E-05	0	0	5944	0.471557333
6	6017	0.000061	0	0	6017	0.367057333
7	6031	0.000153333	0	2.66667E-05	6031	0.924804444
8	6154	0.000101333	0	0	6154	0.623639111
9	6091	2.46667E-05	0	0	6091	0.150236444
10	6066	4.76667E-05	0	0	6066	0.289161889
11	6061	1.06667E-05	0	0	6061	0.064654222
12	6135	0.000032	0	0.00006	6135	0.196309333
13	6012	5.63333E-05	0	0.00002	6012	0.338694778
14	5581	0.000079	0	0	5581	0.440925333
15	6154	5.26667E-05	0	0	6154	0.324110667
16	6248	4.63333E-05	0	0	6248	0.289475222
17	6497	2.86667E-05	0	0	6497	0.186256889
18	6708	0.000049	0	0	6708	0.328708333
19	6630	0.000032	0	0	6630	0.212149333
20	6311	0.000121333	0	0	6311	0.765775111
21	6390	0.000106	0	0	6390	0.677304667
22	6836	5.93333E-05	0	0	6836	0.405582889
23	7184	0.000024	0	0	7184	0.172408
24	6191	8.33333E-06	0	0	6191	0.051588889
25	6228	0.000046	0	0	6228	0.286488
26	6443	5.46667E-05	0	0	6443	0.352235556
27	6527	0.000096	0	0	6527	0.62656
28	6502	8.33333E-05	0	0	6502	0.541861111
29	6385	0.000158	0	0	6385	1.008777333
30	6625	5.53333E-05	0	0	6625	0.366583333
31	6820	0.000884	0	0	6820	6.028585333
Total (scf)	195061		0		195061	18.58378822
Avg (scf/day)	6292		0		6292	9.5272E-05
Volume UF6 :	0.186	ft^3				

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5.5 Calculation of the Isotopic Activity and Mass of ^{234}U , ^{235}U , and ^{238}U

An estimate of the contribution to the total "effective" activity of each uranium isotopic constituent is calculated according to Eq 5.5-1.

$$A_i = f_i S \quad (5.5-1)$$

where f_i is the fraction of total activity for each isotopic constituent from Fig 1-2, and S is the specific activity from Eq 1-2.

The fraction of total "effective" activity, f_i , for each uranium isotope is read from the graph in Fig 1-2 at the assumed 93% ^{235}U enrichment. The accompanying table at the right details the fractions used for each isotope in this calculation. For March 1961, the contribution to the total "effective" activity by ^{234}U is

i	f_i
^{238}U	0.00035
^{235}U	0.031
^{234}U	0.96865

$$A = (0.96865)(3.422 \times 10^{-3}) = 3.314 \times 10^{-3} \text{ Ci } ^{234}\text{U}.$$

The contributions by ^{235}U and ^{238}U are calculated similarly. The estimate of the mass of each uranium isotopic constituent is calculated using equation Eq 1-3 and uses the theoretical specific activities for each uranium isotope computed in Section 5.4. For March 1961, the mass of ^{234}U is calculated as

$$m = \frac{3.314 \times 10^{-3}}{6.2092 \times 10^{-3}} = 5.338 \times 10^{-1} \text{ g } ^{234}\text{U}.$$

The mass of ^{235}U and ^{238}U are calculated similarly. The masses of the three isotopes were summed and compared with the total uranium mass calculated in Section 5.3 to ensure close agreement. The ratio of the ^{235}U mass to the total U mass was computed to ensure an approximate 93% enrichment level. The activity fractions in the table above represent the values resulting from several iterative refinements. The final fraction values from Fig 1-2 used in this calculation result in an overstatement of the total uranium mass by 1.28% over the estimate resulting from the van der Waals real gas equation. The uncertainties in the experimental data and the fitted equation in Fig 1-1 and in selecting values from the graph in Fig 1-2 contribute to the differences in the mass calculations. Also notable is the omission of the ^{234}U contribution to the U and UF_6 molecular weight calculations in Section 5.1 which impact the van der Waals real gas equation calculations in Section 5.3, the specific activity calculations in Section 5.4, and the mass calculations in Section 5.5.

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5.6 Discussion of the Results of the Calculation

This calculation estimates that between March 1961 and December 1961, about 1.545 kg of highly enriched uranium was vented to the atmosphere by the purge cascade. The monthly median during these ten months was about 122 g uranium with a standard deviation of about 102 g uranium. This release occurred daily and constituted a total of about 100 mCi with a monthly median of about 8 mCi and a standard deviation of about 6.6 mCi.

Gaseous diffusion causes a greater percent increase in ^{234}U than ^{235}U due to the better separation factor for ^{234}U . The half-life of ^{234}U is four and five orders of magnitude shorter than ^{235}U and ^{238}U and thus constitutes a higher percent of the overall activity. At 93% ^{235}U enrichment, about 96.9% of the total activity is due to the presence of $^{234}\text{UF}_6$ even though the weight percent of ^{234}U at this enrichment is only about 1%.

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7. ELECTRONIC FILES

The following files are included on the diskette that accompanies this calculation.

<u>File Name</u>	<u>Description</u>
95011R0.DOC	This document (Microsoft® Word version 6.0a).
95011R0.XLS	Spreadsheet for the calculation of the ORGDP purge cascade uranium release estimates for March 1961 through December 1961 (Microsoft® Excel version 5.0a).
RAWPURGE.XLS	Spreadsheet for the raw purge rate data from the K-25 Area 5 Foreman's Logbooks (Microsoft® Excel version 5.0a).

Attachment 1

**Purge Rates February 1961 (partial) - December 1961
(from the K-25 Area 5 Foreman Logbooks at K-1034-A Site Records)**

MONTH MAY 1964

DAY	CELL K-312-1						CELL K-312-2					
	SHIFT 1		SHIFT 2		SHIFT 3		SHIFT 1		SHIFT 2		SHIFT 3	
	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC
1	7099	000015	7191	000078	6992	000015	—	NIL	—	NIL	—	NIL
2	7038	000005	7053	000020	6992	000138	—	"	—	"	—	000020
3	6977	000025	7252	000118	7038	000114	000010	000010	—	"	—	000020
4	7222	000045	7497	000106	7225	000415	—	000200	—	000210	—	000150
5	7803	000060	7359	000087	6962	000051	—	NIL	—	NIL	—	NIL
6	6916	000119	7053	000025	7145	nil	—	"	—		—	
7	7252	000113	7359	000214	7237	000028	—	"	—		—	
8	7206	000145	7344	000070	7145	000019	—	"	—		—	
9	7222	000054	7115	000005	7237	nil	—	"	—		—	
10	7313	000212	7941	000750	7472	000139	—	"	—		—	↓
11	7344	000286	7237	nil	7466	000006	—	000220	—		—	000060
12	7451	000010	7291	000670	7481	000127	—	00010	—		—	000210
13	7426	000070	7482	000220	7421	000020	—	000210	—		—	000120
14	6946	000270	6273	000300	6549	000312	—	000210	—		—	NIL
15	6457	000165	6395	000135	6518	000349	—	NIL	—		—	↓
16	6717	000583	6778	000067	6916	000166	—	"	—		—	↓
17	6732	000025	7069	000319	6732	000074	—	"	—		—	000450
18	6441	000007	6350	000160	6564	nil	—	000450	—	↓	—	000390
19	6656	000030	6885	000018	6931	000385	—	000390	—	000350	—	000410
20	6962	000025	6992	000016	6946	000015	—	000420	—	000490	—	000450
21	7053	000037	6992	000038	6931	000030	—	000430	—	000930	—	000430
22	6977	000051	7160	000225	6922	000515	—	000260	—	000930	—	000290
23	6992	000205	7650	000026	7023	000040	—	000430	—	NIL	—	000220
24	6931	000049	7850	000138	6916	nil	—	000220	—	000290	—	000420
25	6809	nil	7700	000021	6686	000089	—	nil	—	nil	—	000220
26	6793	000012	7825	000031	6977	000059	—	000050	—	"	—	000460
27	7023	000010	6747	000450	6977	000069	—	000340	—	000220	—	nil
28	6809	000015	8000	000060	7130	000121	—	000140	—	nil	—	nil
29	6870	000040	8150	000040	6885	000148	—	000340	—	000220	—	"
30	6839	000075	6931	000081	6930	000130	—	000480	—	000200	—	"
31	6931	000040	6686	000031	6854	000095	—	nil	—	000070	—	"

MONTH JUN 1961

JE Bullman 10/27/95

CELL 312-1

CELL _____

[illegible]

MONTH JUL 1961

282

CELL 312-1							CELL _____					
AY	SHIFT 1		SHIFT 2		SHIFT 3		SHIFT 1		SHIFT 2		SHIFT 3	
	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC
1	7711	000043	8140	00008	7816	000098						
2	8850	000115	8017	000134	7726	000028						
3	7635	000064	7375	000013	7543	000031						
4	7574	000196	8200	000015	7314	000018						
5	7405	000087	7650	000015	7451	000011						
6	7237	000094	6518	000028	6640	00002						
7	6656	000153	7300	000003	6625	000093						
8	6625	000126	7200	000025	6717	000080						
9	6563	000034	7000	000001	6288	000037						
10	6579	000053	7600	000003	6380	000012						
11	6243	000013	6212	000047	6181	000093						
12	6242	000145	6197	000061	6181	000116						
13	6212	000137	6334	000539	6600	000009						
14	6090	000079	6044	000085	6700	000033						
15	6074	000222	8919	000037	6500	000002						
16	6028	000064	6013	000076	6273	000418						
17	5998	000003	6166	000413	5840	000019						
18	5998	000003	5692	000196	6074	000005						
19	6105	000085	6304	000334	7023	000145						
20	7084	00018	6900	00016	7390	00044						
21	5737	000008	7283	000218	7328	000225						
22	7497	000004	7314	000289	7252	000127						
23	7130	000450	7329	000168	7283	000009						
24	7405	000004	7390	000102	7512	000086						
25	7284	00055	7788	000371	7176	000076						
26	7222	000165	7460	000350	7268	000094						
27	7405	000015	7313	000074	7314	000023						
28	7283	NIL	7268	000011	7191	000003						
29	7405	000069	7313	000063	7375	000061						
30	7130	000015	7206	000022	7191	000121						
31	7375	000087	7396	000147	7497	000208						

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[illegible]

①

END OF AUG 1961 } FIRST PART
OF SEPT 1961

10/23/55
JE (Buckley-Sum)

JP

KP-2222 Date of Document 8/21/61

"K-25 Area

Foreman's Logbook" — Page Consider date for OHTS

~~8/17/61~~

~~8/21/61~~

SHIFT		K-312-1		K-311-1	
8/22	4-12	8500 ✓ S.C.D.	0.00034 ✓ $\frac{1}{16}$	17,000 ✓	0.00004 ✓
		10,300 (max)	0.0001	24,000 (max)	0.00004
8/22	12-8	8,400 ✓	0.000051 ✓	20,125 ✓	0.00003
		N/A (max)	0.00000	28,500 (max)	N/A (From 302-4.4)
	8-4	8,231	0.000046 ✓	17,000 ✓	0.00002
		10,300 (max)	0.000146	28,500 (max)	N/A (From 413)

~~8/22/61~~

SHIFT		K-312-1		K-311-1	
	4-12	8400 ✓	0.00017 ✓	17,000 ✓	0.00004 ✓
		10,000 (max)	0.000516	24,000	N/A
8/23	12-8	8155 ✓	0.000041 ✓	18,125 ✓	0.00003
		N/A (max)	0.000151	20,000	N/A (3)
	8-4	8476 ✓	0.00003 ✓	17,000 ✓	0.00004 ✓
		N/A (max)	0.00003 N/A	28,500	N/A (From 12-27)

~~8/23/61~~

	4-12	8415 ✓	0.00007 ✓	17,000 ✓	0.00002
		9900 (max)	N/A	27,500	0.00012 (K-22)
8/24	12-8	8415 ✓	0.000041 ✓	17,000 ✓	0.00002
		N/A	0.000124	18,500	N/A
	8-4	8415 ✓	0.00007 ✓	17,000 ✓	0.00017
		N/A	N/A	18,000	NA (K-33)

689

(2)

SH, FT

1961

K-312-1

PR

TL

K-311-1

PR

TL

8/24 4-12

8400 ✓

0.000029 ✓

NA ✓

NA ✓

9500 (MAX)

0.000081

NA

NA

8/25 12-8

8889 ✓

0.000025 ✓

19,850 ✓

0.00005

NA

NA

26,500

NA (USS)

8-4

8568 ✓

0.000127 ✓

17,000 ✓

0.00014

NA

0.000665

25,700

NA (USS)

4-12

8354

0.000048 ✓

17,000 ✓

NA ✓

9500 max, etc

0.000175

25,000

NA

8/26 12-8

8293 ✓

0.000058 ✓

17,000 ✓

0.00004

10,000

0.00018 From K-413

24,000

0.00012 (K-413)

8-4

8629 ✓

0.00006 ✓

17,000 ✓

0.00018 ✓

NA

0.00009

22,000

NA (K-33)

4-12

8492 ✓

0.000065 ✓

17,000 ✓

NA ✓

18,200

0.000215

28,500

NA

8/27 12-8

NA ✓

NA ✓

17,000 ✓

0.00014 ✓

NA

NA

28,500

0.00014 311-1.5

8-4

8583 ✓

0.000052 ✓

17,000 ✓

0.00014 ✓

NA

0.000095

25,500

NA

4-12

8553 ✓

0.000051 ✓

17,000 ✓

NA ✓

10,100

0.000014

30,000

NA

8/28 12-8

8293 ✓

0.00005 ✓

17,000 ✓

0.00008

7600

0.00006

24,500

0.00024

8-4

8124 ✓

0.000004 ✓

17,000 ✓

0.00014 ✓

NA

NA

24,500

NA

4-12

8446 ✓

0.000078 ✓

16,500

NA

10,200

0.000255

38,500

NA

Q82

③
SHIFT

K-312-1

K-311-1

1961

8/29	12-8	8629 ✓	0.00008 —	17,000 —	0.00004 —
		9900	0.000225	29,000	0.00016
	8-4	8706 ✓	0.000345 —	22,250 ✓	0.00003 —
		NA	NA	62,000	NA (CSI)
	4-12	8981 ✓	0.000254 —	17,000 ✓	0.00003
		12,000	0.000745	65,000	NA (LC-33)
8/30	12-8	8568 ✓	0.00005 —	17,000 ✓	0.00008
		12,000	0.000305	32,500	0.00014
	8-4	9400 ✓	0.000045 —	17,000 ✓	0.00002 —
		9500	0.0006	78,000	0.00002 3013.2
	4-12	8721 ✓	0.000124 —	17,000 ✓	0.000014
		10,800	NA	24,000	NA
8/31	12-8	8492 ✓	0.00005 —	17,000 ✓	0.00004
		NA	0.000175	17,500	0.00016 B-9
	8-4	8767 ✓	0.00017 —	17,000 ✓	0.00002 —
		10,300	0.00044	17,000	0.00002
	4-12	8262 ✓	0.000119	17,000 ✓	0.00013 —
		7700	0.00004	24,000	NA
		NA			
		4700			
9/1	12-8	8446 ✓	0.000093 —	17,000 ✓	NA —
		11,200	0.000365	17,250	0.00015
	8-4	8507	0.000095 —	18,250 —	0.00003 —
		NA	0.000104	NA	NA
	4-12	8293 —	0.000069 ✓	17,000 —	0.00015 —
		NA	0.00014	34,500	NA
9/2	12-8	8921 ✓	0.000164	17,000	NA —
		12,100	0.000375	17,500	0.00002

MONTH SEPT 1961

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CELL 312-1							CELL 311-1						
	SHIFT 1		SHIFT 2		SHIFT 3		SHIFT 1		SHIFT 2		SHIFT 3		
	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	
1		SEE OTHER		SHEETS	FOR	8/21	through	9/2					→
2				→	8629	000107							
3	8507	000023	8323	000005	8736	000005	17000	00002	17000	000003	17000	000013	
4	8553	000010	8293	000005	8384	000007	17000	—	16800	000003	16500	000015	
5	8354	000001	8262	000001	8293	000063	17000	—	17000	B?	17000	000017	
6	8553	000411	8522	000083	8415	NIL	17000	—	17000	000002	17000	000015	
7	8461	000026	8109	000104	7497	000002	17000	—	17000	000002	17000	000003	
8	7696	000024	7711	000002	7619	000003	17000	—	17000	000006	17500	000003	
9	7328	000096	7329	000026	9100	000058	17500	—	17000	000006	17000	000003	
10	7497	000048	7512	000054	7344	000085	21000	000021	17000	000016	17000	000016	
11	7405	000004	7329	000058	7268	000025	17500	000012	17500	000004	17000	000003	
12	7053	000034	6992	000003	6962	000002	17000	000015	17000	—	17000	000002	
13	6870	000012	6916	000007	6921	NIL	17000	000001	17500	000001	17000	000003	
14	7084	000015	7700	0000176	8100	000003	17000	000015	17500	—	17000	000002	
15	8100	000005	8500	0000033	7900	0000025	17500	000015	18000	—	17500	000002	
16	7400	000009	8400	000006	8100	000002	17000	—	18000	—	18000	—	
17	8400	000007	8200	0000066	8100	000006	20000	000003	19000	—	18000	000002	
18	7965	000002	8700	0000147	8100	000004	21000	000002	—	—	17500	000008	
19	7560	000001	8375	0000015	7560	000007	18500	000007	18500	18500	18500	—	
20	7496	000001	8300	0000105	7560	000002	17500	000003	—	—	17500	000001	
21	7290	0000065	8200	0000133	8100	0000082	17500	000002	17500	—	18000	—	
22	8288	000013	8325	0000195	8200	0000033	18000	000003	19500	—	18000	—	
23	8200	0000088	8400	000049	8400	—	17500	000008	18000	—	17500	—	
24	8800	000024	8575	0000063	8400	0000093	17500	000006	17500	—	17000	—	
25	8700	NIL	8800	0000059	8500	0000063	17500	000014	17500	—	17500	—	
26	8800	0000122	8675	0000045	8700	0000119	17500	000001	17500	—	17500	—	
27	8650	NIL	5050	000012	8600	0000056	17500	—	18000	—	17700	—	
28	8550	000004	9050	MAINT.	9057	0000045	17500	000001	17500	000001	17500	000013	
29	9300	000003	9175	0000025	9375	—	17500	000008	17500	000003	17500	000015	
30	8800	0000031	8750	000005	9038	—	18200	—	18000	000001	17500	000001	
31	8800	0000031	8750	000005	9038	—	18200	—	18000	000001	17500	000001	

MONTH OCT 1961


882

CELL 312-1							CELL 311-1					
Y	SHIFT 1		SHIFT 2		SHIFT 3		SHIFT 1		SHIFT 2		SHIFT 3	
	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC
1	9000	000027	9400	00003	9413	00003	18200	—	18000	00003	17500	0001
2	9500	000024	8800	NIL	8800	000028	18500	—	18500	00002	18500	00015
3	9400	000032	9400	00003	10287	000036	18500	—	18500	00012	18500	0001
4	9300	000047	9700	00004	9600	000159	19200	—	18000	—	18000	00015
5	9300	0000491	9425	00016	9562	0002	18000	—	16000	00015	16500	0002
6	9825	—	9142	00003	9493	000025	17500	—	16500	00015	16500	00012
7	9331	00014	7340	000058	7020	000015	16500	—	16500	—	16000	0004
8	8750	000038	7460	00008	7220	00013	16500	00015	16000	00002	16000	—
9	8700	000129	7450	00015	7695	NIL	16500	00015	15500	15500	16000	00004
10	9113	00004	8000	NIL	7763	00007	16000	00015	15000	—	15500	00002
11	7789	00209	7800	000097	7965	000125	15500	00475	15500	—	15500	00002
12	7425	00002	7830	000118	7560	00002	15500	00015	15500	—	15500	00002
13	8303	000108	9382	000385	9787	0003	15500	00015	15500	—	15500	0002
14	9248	00008	10,200	000088	8775	000006	15500	00002	16000	—	16000	0001
15	8910	000198	8300	000049	7695	NIL	16000	00002	15500	—	15500	00004
16	7628	NIL	7560	NIL	7897	00021	16000	00019	15500	—	15500	00025
17	8640	000015	8505	000059	9315	000027	15500	—	15500	00013	15500	00005
18	9315	NIL	9113	000205	5675	0001	15500	00012	15500	—	15500	0001
19	6015	000225	76100	000065	6000	000091	16500	00024	15500	00017	16000	—
20	5175	000135	5113	000057	5750	000016	15500	00018	15500	00012	16000	—
21	5035	00005	5500	000249	5000	000036	16000	0001	15500	00013	16500	—
22	5125	000108	5450	000152	5450	00001	15500	00002	15500	00002	15500	—
23	5000	00001	5425	—	5470	0001	16000	00016	16000	00006	16000	—
24	5200	00018	5575	NIL	5800	000094	16000	—	24000	00026	16000	—
25	5250	NIL	7200	000125	6450	000187	16000	00014	17500	00015	15500	—
26	6400	0002	5438	000285	6463	000076	15500	0001	16000	00024	15500	—
27	6125	00002	5500	NIL	5850	000214	15500	00012	16000	00008	—	—
28	6250	000076	5850	NIL	5150	000051	16000	—	16000	00006	15500	00012
29	6000	000195	6625	NIL	5738	000035	15500	—	15500	00002	15500	00001
30	5225	000057	—	—	5413	NIL	16000	—	16000	—	16500	00003
31	5575	NIL	5250	000023	5138	000047	16000	—	16000	00002	16000	00012

MONTH- DEC 1961

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NY	CELL 312-1						CELL 311-1					
	SHIFT 1		SHIFT 2		SHIFT 3		SHIFT 1		SHIFT 2		SHIFT 3	
	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC
1	5800	NIL	6287	NIL	5912	NIL	17000	—	17000	0001	17000	00013
2	5919	000159	5400	NIL	5662	00005	17000	00012	17000	00012	17000	0001
3	5788	000059	5700	NIL	5750	00007	17000	00012	16500	00012	17000	0003
4	5825	000087	5925	NIL	6050	00006	17000	00012	17500	00014	17000	0003
5	5969	000034	6350	NIL	5950	0000154	17000	00013	17500	00014	17500	00009
6	6019	000086	5900	—	5925	000029	17500	00013	17000	—	17500	0002
7	6394	0003	6060	000129	5725	00002	17500	00013	17500	—	17500	0001
8	5450	000237	5150	0003	5550	00008	17500	00013	17500	00014	17500	00011
9	5650	00084	5300	00005	6062	00008	17500	0005	17000	—	17000	00004
10	5075	000034	5487	000034	5950	00016	17000	00002	17000	—	17500	00006
11	5375	000079	—	—	5725	00005	17500	00014	17500	—	17000	0001
12	5950	000705	6450	000182	6550	00014	17000	0004	17000	00004	17000	00012
13	6550	000648	6819	000077	6925	000025	17500	0001	17000	—	17000	00012
14	7075	NIL	7700	000126	6900	NIL	17000	00017	17000	00013	17500	—
15	6675	00003	6894	000052	7000	NIL	17500	0002	17500	00012	17500	—
16	6675	000049	6725	000074	6800	000079	17000	0001	17500	00012	17000	—
17	6700	NIL	7028	000024	6400	NIL	17000	00002	17000	00012	17000	—
18	6775	NIL	6838	000043	6500	NIL	17000	00002	17000	00012	17000	—
19	6725	NIL	6800	NIL	6300	NIL	17000	—	17000	00012	17500	—
20	6850	000035	7175	000335	7000	NIL	17000	—	17000	—	17000	—
21	6700	00006	7075	000035	6894	000014	17000	—	17000	00006	17000	00014
22	7100	NIL	6600	NIL	7438	000026	17000	—	17000	0001	17500	00012
23	7000	NIL	7700	000022	7063	000026	17500	—	17500	00002	17000	00012
24	6900	NIL	6700	—	6994	000025	17000	—	17000	0001	17000	00011
25	6800	000442	7025	NIL	7000	000016	16500	—	17000	0001	16500	00011
26	7000	NIL	7000	NIL	7100	000002	16500	—	16500	—	17000	00012
27	6800	—	7200	NIL	6900	000068	17000	22000	17000	—	16500	0001
28	6800	000192	6775	NIL	6975	NIL	17500	—	17000	00012	17000	00003
29	6200	000102	6600	000055	7375	00014	16500	—	17000	00011	17000	00005
30	7187	000034	7175	00013	7125	0001	17000	00012	17000	0001	17000	00005
31	7118	00003	7275	00005	7325	00001	17000	00012	1700	00011	17000	00008

[illegible]

MONTH MARCH 1961

280

DAY	CELL <u>312-1</u>						CELL <u>312-2</u>					
	SHIFT 1		SHIFT 2		SHIFT 3		SHIFT 1		SHIFT 2		SHIFT 3	
	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC
1	5909	000034	6101	000200	5968	000065	—	NIL	—	NIL	—	NIL
2	6101	000058	6174	000105	6189	000036	—	"	—	"	—	"
3	5983	000109	6086	000061	6145	000065	—	"	—	"	—	"
4	5954	000205	6159	000079	6101	000018	—	000040	—	"	—	"
5	5807	000035	5924	000096	6101	000107	—	NIL	—	"	—	"
6	5821	000030	6042	000091	6189	000062	—	"	—	"	—	"
7	5865	000030	6114	000267	6115	000063	—	000080	—	"	—	"
8	6012	000200	6262	000079	6189	000025	—	NIL	—	"	—	"
9	6027	000010	6145	000024	6100	000040	—	"	—	"	—	"
10	5998	000015	6012	000019	6189	000109	—	"	—	"	—	"
11	6233	000014	5880	000010	6071	000008	—	"	—	"	—	"
12	6203	000015	6145	000073	6056	000008	—	000080	—	"	—	"
13	6071	000089	6086	000031	5880	000049	—	000060	—	"	—	"
14	5630	000034	5557	000105	5557	000098	—	NIL	—	"	—	"
15	6321	000036	5967	000044	6174	000078	—	"	—	"	—	"
16	6248	000030	5792	000065	6073	000044	—	"	—	"	—	"
17	6600	000035	6424	000030	6468	000021	—	"	—	"	—	"
18	6733	000123	6277	000010	6615	000014	—	"	—	"	—	"
19	6630	000022	6615	000040	6644	000034	—	"	—	"	—	"
20	557	000014	6115	000080	6262	000272	—	"	—	"	—	"
21	6380	000273	6380	000015	6409	000030	—	"	—	"	—	"
22	6703	000049	6777	000055	7027	000076	—	"	—	"	—	"
23	6983	000015	7159	000057	7409	NIL	—	"	—	"	—	"
24	6571	000005	6048	000001	5953	000010	—	"	—	"	—	"
25	6189	000025	6189	000053	6306	000060	—	"	—	"	—	"
26	6468	000016	6497	000048	6365	000100	—	"	—	"	—	"
27	6615	000110	6512	000078	6453	000100	—	"	—	"	—	"
28	6497	000051	6424	000024	6586	000125	—	"	—	"	—	"
29	6350	000038	6365	000066	6439	000320	—	"	—	"	—	"
30	6512	000028	6630	000062	6733	000026	—	"	—	"	—	"
31	6512	000224	6895	000223	6262	000205	—	"	—	"	—	"

[illegible]

DSF #3189

SHONKA RESEARCH ASSOCIATES, INC.

Calculation Control Sheet

Calculation number: SRA-95-013 REV. 0

Title: URANIUM RELEASE ESTIMATES FOR THE ORGDP
PURGE CASCADE 7/75 - 6/76

Reason for calculation/revision: ORIGINAL

Client: ATOMIC RISK, INC.

Project: OAK RIDGE HEALTH STUDIES

Project/Task Number: TASK 6

Prepared by: TE BUNNETT Date: 11/10/95

Independent Technical Review by: *[Signature]*

Date: 12/1/95

Quality Assurance Review by: Deborah B. Shonka

Date: 12/4/95

☐ This calculation has been voided or superseded by _____
(calculation number)

This document has been reviewed for classification and has been determined to be UNCLASSIFIED.	
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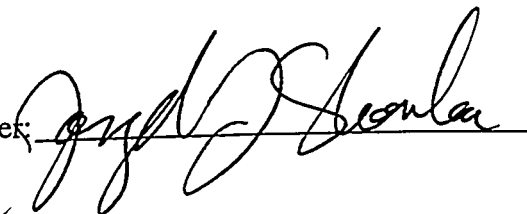
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Review Method Sheet

The undersigned has reviewed this calculation in accordance with the method(s) indicated below.

1. Computer Aided Calculation	
a	Review to determine that the computer program(s) has been validated and documented, is suitable to the problem being analyzed, and that the calculation contains all necessary information for reconstruction at a later date.
b	Review to determine that the input data as specified for program execution is consistent with the design input, correctly defines the problem for the computer algorithm and is sufficiently accurate to produce results within any numerical limitations of the program.
c	Review to verify that the results obtained from the program are correct and within stated assumptions and limitations of the program and are consistent with the input.
d	Review validation documentation for temporary changes to listed, or developmental, or unique single application programs, to assure that the methods used adequately validate the program for the intended application.
e	Review of code input only, since the computer program has sufficient history of use at Shonka Research Associates, Inc. in similar calculations.
f	Review arithmetic necessary to prepare code input data.
g	Other:
2. Hand Prepared Calculations	
a	Detailed review of the original calculations.
b	Review by an alternate, simplified, or approximate method of calculation.
c	Review of a representative sample of repetitive calculations.
d	Review of the calculation against a similar calculation previously performed.
e	Other:
3. Revisions	
a	Editorial changes only
b	Elimination of unapproved input data without altering calculated results.
c	Other:
4. Other	

Reviewer:



Date: 12/1/95

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12/11/95
J. J. ShonkaTitle Uranium Release Estimates for the ORGDP Purge Cascade 7/75 - 6/76

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ABSTRACT

This calculation represents the fifth in a series of calculations aimed at establishing a methodology and strategy in analyzing the quality of the historical effluent monitoring data and practices at the Oak Ridge Gaseous Diffusion Plant. Within that scope, the emission rates from the purge cascade during the months of July 1975 through June 1976 were analyzed in order to determine its magnitude as a site contributor in the total historical uranium emissions. Daily purge rate data that documented the volumetric flow of the purge gas and its concentration of UF_6 were used to compute the daily flow of UF_6 in the purge cascade. The total volumetric flow of UF_6 for each month was used in the modified van der Waals real gas equation in order to estimate the mass of UF_6 released. Calculations show that over the one year period, about 210 g of slightly enriched UF_6 was measured by the space recorder monitoring instrumentation located in cell K-311-1 of the purge cascade. This corresponds to the release of about 252 μCi of alpha activity and includes contributions from the mixture of $^{234}\text{UF}_6$, $^{235}\text{UF}_6$, and $^{238}\text{UF}_6$. These estimates do not include any reductions due to carbon or alumina traps located downstream of the monitoring instrumentation prior to the purge stack exit.

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Attachments

1. Purge Rates July 1975 - June 1976 (from the K-25 Area 5 Foreman Logbooks at K-1034-A Site Records)

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1. INTRODUCTION

Task 6 of the Oak Ridge Dose Reconstruction focuses on the evaluation of the quality of historical airborne and waterborne effluent monitoring data and the determination of the potential significance of unmonitored emissions. Uranium played an important role throughout historical operations on the Oak Ridge Reservation (ORR) and is known to have been released to the environment through air and water. The two largest uses of uranium on the Reservation were the enrichment processes of the ^{235}U isotope by electromagnetic separation at the Y-12 facility and gaseous diffusion at the K-25 facility.

Task 6 activities will be directed at establishing revised uranium release estimates with an associated uncertainty over that of the screening analyses conducted during the Dose Reconstruction Feasibility Study. These activities will support refined assessment of the potential magnitude of health hazards from historical uranium exposures based on both the chemical and radiotoxicity of uranium.

This calculation represents the fifth in a series of calculations aimed at establishing a methodology and strategy in analyzing the quality of the historical effluent monitoring data and practices at the Oak Ridge Gaseous Diffusion Plant (ORGDP). For many years the purge cascade represented the only on-site monitored emission source. This along with previous and subsequent calculations will provide a historical review of the uranium released from the ORGDP purge cascade including an assessment of the potential uncertainties and biases in the measurement and computation of the estimates.

Light molecular weight gases were purged from the top of the diffusion cascade that would otherwise block the withdrawal of enriched UF_6 product. These light gases originated from the following sources.

- *nitrogen* - mainly from the inleakage occurring at every pump shaft seal in the diffusion cascade
- *hydrogen fluoride* - from reaction of inleaking moist air with UF_6
- *oxygen, argon* - from inleaking air
- *chlorine fluorides* - used in conditioning and drying of metal surfaces
- *fluorine* - used in conditioning of metal surfaces
- *coolant vapor* - inleakage from the compressor and pump coolant system

The light gases in the process stream had a molecular weight substantially less than that of the UF_6 component and were carried along effectively by the diffusion process to the top of the cascade. A section at the top of the cascade just above the product withdrawal point was reserved as a the purge cascade. The purge cascade separated these light gases from the enriched UF_6 product and vented them to the atmosphere. The purge cascade usually existed in two sections: a side purge and a top

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purge. The side purge separated the intermediate molecular weight gases (*i.e.* coolant vapor, chlorine fluorides, etc.) from the UF_6 . The top purge separated the remaining lighter gases. At various times throughout the operational history, the side purge was routed to the top purge. The effluents were pumped through traps in order to reduce the uranium content before venting to the atmosphere. In the earlier years these traps consisted only of carbon and alumina, but later sodium fluoride (NaF) traps and potassium hydroxide (KOH) scrubbers were added. Because of the large difference in molecular weights between the light gases and UF_6 , only a few diffusion stages were needed to effectively perform the separation. Similar to the main diffusion cascade, the purge cascade consisted of compressors, converters, motors, coolers, piping, control and block valves, and instrumentation. The major difference between the main diffusion cascade and the purge cascade was the smaller amount of UF_6 flow (MMES 1985).

The problem of analyzing the gas in the purge cascade was complicated by the fact that the UF_6 concentration varied greatly from one end of the cascade to the other. Near the bottom of the cascade the process stream consisted of essentially pure UF_6 , whereas at the cascade top the stream consisted of light gases containing only small traces of UF_6 . The line recorder was designed for analyzing UF_6 containing relatively small amounts of impurities. The method it employed measured the flow of gas to a mass spectrometer tube by means of a Pirani gauge flowmeter; the UF_6 was removed chemically before reaching the spectrometer tube and the residual gas concentration measured by means of the spectrometer tube. In analyzing for UF_6 in the presence of relatively large amounts of impurities, the accuracy dropped sharply. Due to the corrosive nature of UF_6 , the concentration could not be directly measured by the spectrometer tube. Therefore, the UF_6 concentration was determined by the difference between the flow computed from the flowmeter and the remaining light gas concentration determined by the mass spectrometer tube reading. Only with careful calibration was it possible to determine either of these quantities with an accuracy of 1% or better. Accordingly, the line recorder became practically useless for determining the composition of a mixture containing under 2% of UF_6 (OM-48 1945).

In order to supplement the line recorder in the purge cascade, an instrument known as the space recorder was developed. The principal component of the space recorder was an ionization chamber more commonly referred to as the "signal can." The signal can measures the specific radioactivity of the gas present, and since UF_6 is an alpha emitter, this method provided a convenient means for measuring the UF_6 content of gas samples. The space recorder could detect the presence of mol fractions of UF_6 in the light gas purge of the order of 10^{-6} (OM-48 1945). The radioactivity of UF_6 consists of the emission of high energy alpha particles at a definite uniform rate. This rate depends upon the relative isotopic composition, since all three isotopes emit alpha particles at a different rate. These alpha particles as emitted have a definite range of travel, which is inversely proportional to the pressure. In the gas samples, at standard temperature and pressure, this range of travel is on average approximately 3 cm. While travelling this distance the energy of the emitted particle is expended by the production of about 130,000 ions resulting from collision of the particles with gas molecules.

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The collection of these ions and a measurement of the resultant electrical current constitutes a determination of the number of particles present and hence a determination of the UF_6 concentration. A record of the purge gas volumetric flow and the concentration of UF_6 in the purge stream during the 1975-1976 time frame was maintained in the Area 5 Foreman Logbook.

This calculation analyzes a one year span of daily purge cascade effluent data for the months of July 1975 through June 1976. These estimates do not represent an actual estimation of the amount of uranium released to the atmosphere during this time period. The sample withdrawal point for the space recorders were located upstream of the carbon and alumina traps. The traps would have removed much of the uranium prior to the venting of the purge gases to the atmosphere and these benefits are not included in these estimates. With this limitation in mind, this calculation makes liberal use of the terms "release", "released", "vented", and "effluent." Furthermore, these estimations are also subject to certain assumptions, biases, and other uncertainties. Several assumptions and potential sources of uncertainty will be presented in this calculation, but the analyses to quantify the impact of these assumptions to the results of this calculation will be formalized in subsequent calculations.

In this calculation, data sheets containing the record of the daily purge rates for the months of July 1975 through June 1976 were transcribed from the logbooks maintained by the Area 5 Foreman and transferred to spreadsheets (Purge Rates 1976). The volume of gas purged each day and its UF_6 concentration was used to compute the daily volumetric flow of UF_6 released. The daily flow of UF_6 was summed to compute the estimate of the total volume of UF_6 vented during the month. The mass of UF_6 released each month in the purge cascade was derived from this known volume at standard conditions by application of the modified van der Waals real gas equation (Ackley, Magnuson 1951) as given in Eq. 1-1,

$$m = \frac{P(1 + AP)V}{RT} \quad (1-1)$$

where P is the pressure of the gas,

A is the temperature-dependent van der Waals coefficient for UF_6 ,

V is the volume of the gas,

R is the UF_6 gas constant, and

T is the temperature of the gas.

The activity of UF_6 released each month in the purge cascade was computed by multiplying the grams of UF_6 by the specific activity of UF_6 at the assumed ^{235}U enrichment level. The "effective" specific activity of a mixture of $^{234}\text{UF}_6$, $^{235}\text{UF}_6$, and $^{238}\text{UF}_6$ (as found in the purge cascade effluent) follows Eq. 1-2.

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$$S = (0.4 + 0.38E + 0.0034E^2) \times 10^{-6} \text{ Ci/g} \quad (1-2)$$

where E is the percent ^{235}U by weight. Eq. 1-2 is fitted to the experimental data in Fig 1-1 (Rich 1988). Eq. 1-2 is fitted to the experimental data in Fig 1-1 (Rich 1988). The contribution to the total "effective" activity of each isotope of uranium was determined from the graph presented in Fig 1-2 (Rich 1988) and used to determine the activity of each isotope. The mass of ^{234}U , ^{235}U , and ^{238}U in the purge effluent could then be calculated from its activity and theoretical specific activity as given by Eq. 1-3. The results of the isotopic mass calculations were compared to the mass calculations for UF_6 using Eq. 1-1 in order to determine the "goodness" of the values selected from Fig 1-2.

$$m_i = \frac{A_i}{S_i} \quad (1-3)$$

where A_i is the activity of the radioisotope, and
 S_i is the specific activity of the radioisotope.

The theoretical specific activity of each uranium isotope is calculated by Eq. 1-4.

$$S_i = \frac{\lambda_i N_A}{M_i} \quad (1-4)$$

where λ_i is the decay constant of the radioisotope,
 N_A is Avogadro's Number, and
 M_i is the atomic weight of the radioisotope

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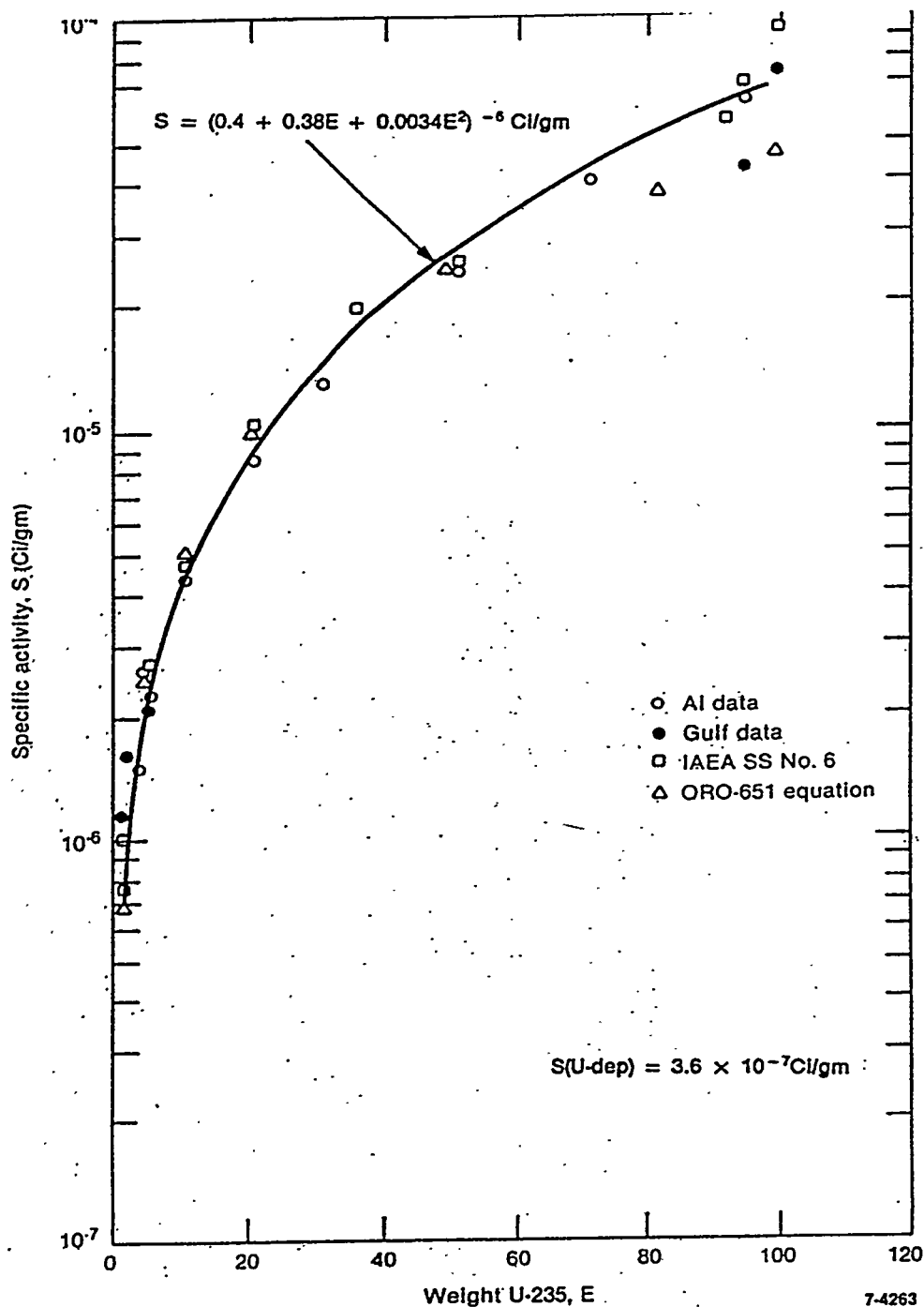


Fig 1- 1 : Specific Activity for Mixtures of ^{234}U , ^{235}U , and ^{238}U

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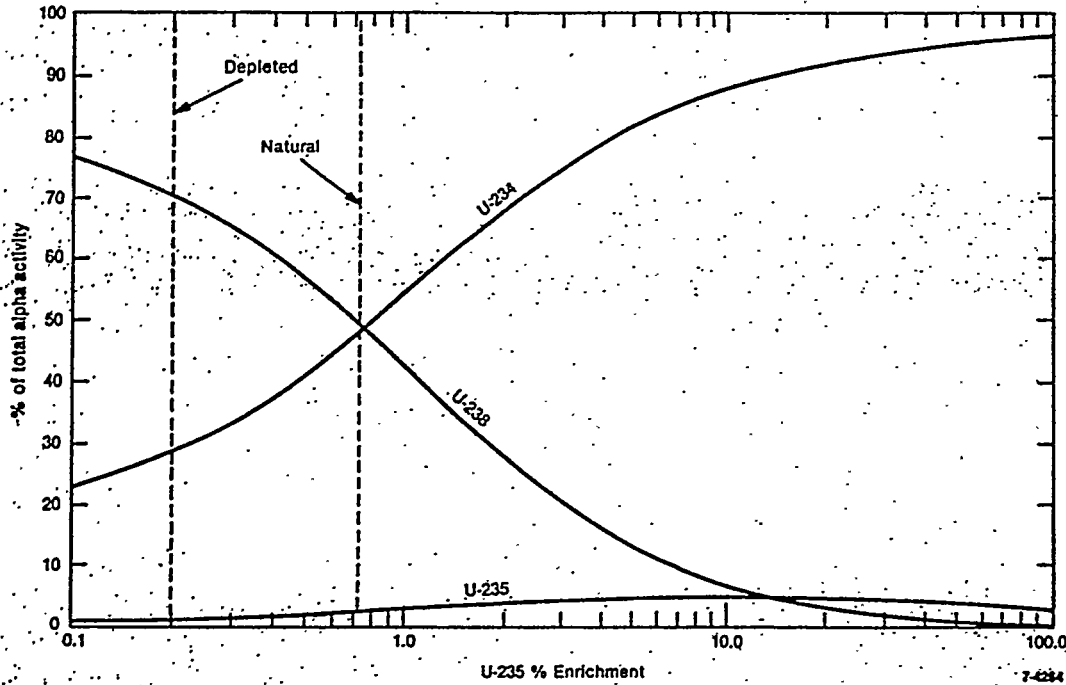


Fig 1-2 : Percent of total radioactivity by isotope vs. % weight ^{235}U enrichment

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2. SUMMARY OF RESULTS

The monthly uranium release estimates from the ORGDP purge unit between July 1975 and June 1976 are presented below in Table 2-1. Totals are given for sum of all months. In addition, the median and standard deviation are given for all computations. The first column of data presents the results of the UF_6 mass calculations using the modified van der Waals real gas equation in Eq. 1-1. The mass of the F_6 is subtracted from the UF_6 mass and the results presented in the second column along with the computation of the "effective" activity given by Eq. 1-2. The next three columns of data present the mass and activity of ^{238}U , ^{235}U , and ^{234}U respectively. The activity of each of the three constituents are computed by applying the appropriate activity fraction from Fig 1-2 to the "effective" activity. The mass of each of the three constituents are computed using the theoretical specific activities of each radioisotope. The last column presents the sum of the masses of each of the three constituents and the percent difference with the uranium mass based upon the modified van der Waals real gas equation computation.

The total UF_6 release over the one year period is 210.1 g with 142.1 g being uranium. This corresponds to a total "effective" activity of 251.7 μCi and assumes a 3.5% ^{235}U enrichment. The "effective" activity corresponds to alpha decay contributions from the mixture of ^{234}U , ^{235}U , and ^{238}U . According to Fig 1-2 at 3.5% ^{235}U enrichment, a majority of the activity is due to the presence of ^{234}U . At this assumed enrichment, about 77.4% of the activity results from the ^{234}U isotope, 18.3% from the ^{238}U isotope, and the remaining from the ^{235}U isotope. The results also indicate that at this assumed ^{235}U enrichment, the ^{234}U isotope is enriched from 0.0056% natural abundance to just over 0.022% and the ^{238}U isotope is depleted to about 96.5%. The sum of the three isotopic masses overstate the calculated uranium mass from the van der Waals gas equation by 0.05%. This difference is subject to uncertainty in the experimental data and the fitted equation in Fig 1-1 and to uncertainty in reading the data from the graph in Fig 1-2.

These estimates do not include any reductions due to carbon or alumina traps located downstream of the space recorder instrumentation prior to the purge stack exit.

Table 2-1: Purge Cascade Uranium F_6 Release Estimates Jul 1975 - Jun 1976

	UF6	U		U-238		U-235		U-234		Total	
Month	[g]	[g]	[Ci]	[g]	[Ci]	[g]	[Ci]	[g]	[Ci]	[g]	Δ%
Jul-75	1.431E+01	9.672E+00	1.714E-05	9.334E+00	3.136E-05	3.410E-01	7.368E-07	2.136E-03	1.326E-05	9.677E+00	0.05%
Aug-75	1.602E+01	1.083E+01	1.919E-05	1.046E+01	3.513E-05	3.820E-01	8.254E-07	2.393E-03	1.486E-05	1.084E+01	0.05%
Sep-75	1.242E+01	8.397E+00	1.488E-05	8.104E+00	2.723E-05	2.961E-01	6.397E-07	1.854E-03	1.151E-05	8.402E+00	0.05%
Oct-75	1.351E+01	9.136E+00	1.619E-05	8.816E+00	2.962E-05	3.221E-01	6.960E-07	2.018E-03	1.253E-05	9.141E+00	0.05%
Nov-75	1.514E+01	1.023E+01	1.813E-05	9.876E+00	3.318E-05	3.609E-01	7.796E-07	2.260E-03	1.403E-05	1.024E+01	0.05%
Dec-75	1.669E+01	1.128E+01	1.999E-05	1.089E+01	3.659E-05	3.979E-01	8.597E-07	2.492E-03	1.547E-05	1.129E+01	0.05%
Jan-76	1.871E+01	1.265E+01	2.241E-05	1.221E+01	4.101E-05	4.461E-01	9.637E-07	2.794E-03	1.735E-05	1.266E+01	0.05%
Feb-76	1.734E+01	1.173E+01	2.077E-05	1.132E+01	3.802E-05	4.135E-01	8.933E-07	2.589E-03	1.608E-05	1.173E+01	0.05%
Mar-76	1.800E+01	1.217E+01	2.156E-05	1.175E+01	3.946E-05	4.292E-01	9.272E-07	2.688E-03	1.669E-05	1.218E+01	0.05%
Apr-76	2.799E+01	1.893E+01	3.353E-05	1.827E+01	6.136E-05	6.674E-01	1.442E-05	4.180E-03	2.595E-05	1.894E+01	0.05%
May-76	2.022E+01	1.367E+01	2.423E-05	1.320E+01	4.433E-05	4.822E-01	1.042E-05	3.020E-03	1.875E-05	1.368E+01	0.05%
Jun-76	1.977E+01	1.336E+01	2.368E-05	1.290E+01	4.333E-05	4.713E-01	1.018E-05	2.952E-03	1.833E-05	1.337E+01	0.05%
Total	2.101E+02	1.421E+02	2.517E-04	1.371E+02	4.606E-05	5.010E+00	1.082E-05	3.138E-02	1.948E-04	1.421E+02	0.05%
Median	1.702E+01	1.151E+01	2.038E-05	1.110E+01	3.730E-05	4.057E-01	8.765E-07	2.541E-03	1.578E-05		
Std Dev	3.924E+00	2.653E+00	4.700E-06	2.560E+00	8.601E-07	9.355E-02	2.021E-07	5.859E-04	3.638E-06		

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3. METHODS

The results summarized in Table 2-1 were based upon data sheets generated from the K-25 Area 5 Foreman Logbooks. These data sheets contain the log of the purge unit volumetric flow and UF_6 concentration for each of the three shifts as measured at purge cell K-311-1, and are included in this calculation as Attachment 1. As part of this calculation, these data sheets were recreated as individual spreadsheets in a Microsoft® Excel workbook and are provided electronically in file 95013R0.XLS. In order to determine an estimate for the total amount of uranium released by the purge unit over this one year period, a number of computations using the raw purge data were required.

During the 1975-1976 time frame, the purge unit consisted of a single cell, K-311-1, operating as both top and side purge. The raw purge data for each day in the Area 5 Foreman Logbooks consisted of measurements recorded during the night, day, and evening shifts. This three shift purge data must first be averaged to daily purge flows. These daily purge gas flows were subsequently summed to compute the total purge gas flow for the month and divided by the number of days in the month to compute the average daily purge gas flow.

Likewise, the raw purge data for each day in the Area 5 Foreman Logbooks consisted of measurements of the UF_6 concentration in the purge gas during the night, day, and evening shifts. This three shift UF_6 concentration data must first be averaged to daily UF_6 concentrations for the purge unit. The daily UF_6 volumetric flow was calculated by multiplying the UF_6 concentration and the purge gas volumetric flow. These daily UF_6 flows were summed to compute the total UF_6 volume released by the purge cascade for the month.

The mass of UF_6 released by the purge unit was calculated using Eq 1-1 and the monthly total volume of UF_6 , the modified van der Waals pressure, standard temperature, and the UF_6 gas constant. The UF_6 mass was multiplied by the ratio of the molecular weight of uranium to the molecular weight of UF_6 resulting in the computation of the uranium mass. The "effective" specific activity of uranium at the assumed 3.5% ^{235}U enrichment was calculated using Eq 1-2 and the result multiplied by the uranium mass to compute the "effective" activity of the uranium. The fractional contribution of this "effective" activity by ^{234}U , ^{235}U , and ^{238}U are obtained from the graph in Fig 1-2 and each multiplied by the "effective" activity to compute the estimated activity of each isotopic constituent. The theoretical specific activity of each constituent was calculated using Eq 1-4 and then divided into the respective activity estimates according to Eq 1-3 to compute the estimated mass of each isotopic constituent. The three masses of the constituents were summed and compared to the uranium mass based upon Eq 1-1 in order to ensure the activity fractions obtained from the graph in Fig 1-2 were appropriate. The activity fraction values were iteratively refined until good agreement between the two mass calculations were obtained.

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4. ASSUMPTIONS

- 4.1. The atomic weight of ^{238}U is given as 238.05077 g/mol (Physics 1967).
- 4.2. The half-life of ^{238}U is given as 4.46×10^9 yr (Physics 1967).
- 4.3. The atomic weight of ^{235}U is given as 235.043915 g/mol (Physics 1967).
- 4.4. The half-life of ^{235}U is given as 7.04×10^8 yr (Physics 1967).
- 4.5. The atomic weight of ^{234}U is given as 234.040904 g/mol (Physics 1967).
- 4.6. The half-life of ^{234}U is given as 2.46×10^5 yr (Physics 1967).
- 4.7. The atomic weight of fluorine is given as 18.998403 g/mol (Physics 1967).
- 4.8. Avogadro's Number is given as 6.022045×10^{23} atoms/mol (Physics 1967).
- 4.9. For purposes of U and UF_6 molecular weight calculations, the contribution due to $^{234}\text{UF}_6$ are assumed negligible.
- 4.10. Standard pressure is assumed 14.7 psia (Lee 1989).
- 4.11. Standard temperature is assumed 59 F or 519 R (Lee 1989).
- 4.12. The universal gas constant \bar{R} is given as $10.73 \text{ psia ft}^3 \text{ lb}^{-1} \text{ mol}^{-1} \text{ R}^{-1}$ (Black, Hartley 1985).
- 4.13. The uranium enrichment of the UF_6 in the purge stream for the purposes of this calculation is assumed to be 3.5% ^{235}U . During the time frame of this calculation, the ORGDP was engaged in process activities producing slightly enriched uranium for the commercial light water reactor power industry. The precise enrichment characteristic of the product varied depending upon customer requirements, but ranged between 2.5% and 5.0% ^{235}U .
- 4.14. The conversion factor of 453.6 g/lb is used to convert between units of mass (Black, Hartley 1985).
- 4.15. The conversion factors of 365.25 days/yr, 24 hr/day, and 3600 s/hr are used to convert between units of time.
- 4.16. The conversion factor of 3.7×10^{10} Bq/Ci is used to convert between units of activity.

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- 4.17. The UF_6 concentrations recorded on the purge rate data sheets are assumed given as mole weight percentages.
- 4.18. Purge gas flow and UF_6 concentration measurements in the K-311-1 cell represent the sum total of the flow in the purge unit.
- 4.19. This calculation assumes UF_6 in the purge stream behaves as a real gas following the behavior prescribed by the modified van der Waals gas equation. The van der Waals coefficient for UF_6 is a function of temperature and has the values of 0.033 atm^{-1} at 141.7 F and 0.021 atm^{-1} at 201.0 F (Ackley, Magnuson 1951).
- 4.20. The trap efficiencies are assumed negligible for the purposes of this calculation, thus implying that the purge gas flow and UF_6 concentration recorded on the purge rate data sheets are identical to the material actually vented to the atmosphere. The impact to the uranium release estimates due to actual trap efficiency, sampling biases and losses, and measurement uncertainties will be addressed in subsequent calculations. Some of the relevant sampling issues are (1) the maintenance of sufficient sample line temperature to prevent UF_6 condensation, (2) losses due to the geometry of sample lines, and (3) measurement uncertainties in the space recorder. The ^{235}U assay will also have an impact on the uranium release estimates.

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5. CALCULATION

5.1 Calculation of the UF₆ Gas Constant

The symbol R in Eq. 1-1 is called the *gas constant* and its value depends upon the particular gas being considered. The value of R for each gas is determined by the equation

$$R = \frac{\bar{R}}{M} \quad (5.1-1)$$

where \bar{R} is a physical constant called the *universal gas constant* and is given in Section 4.3. It is first necessary to compute the atomic weight of uranium. Eq 5.1-2 gives the molecular weight of an isotopic mixture as

$$\frac{1}{M} = \frac{1}{100} \sum \frac{w_i}{M_i} \quad (5.1-2)$$

and for 3.5% ²³⁵U

$$\frac{1}{M} = \frac{1}{100} \left(\frac{3.5}{235.043915} + \frac{96.5}{238.05077} \right),$$

which gives $M = 237.9442315$. The molecular weight of UF₆ is then $237.9442315 + (6)(18.998403) = 351.9346495$. Substituting into Eq. 5.1-1, the value of R for UF₆ becomes

$$R = \frac{10.73}{351.9346495} = 0.030488615 \text{ psia ft}^3 \text{ lb}^{-1} \text{ R}^{-1}.$$

5.2 Calculation of the Modified van der Waals Pressure

Since UF₆ behaves as a real gas, the modified van der Waals pressure is required to account for the non-ideality of the UF₆ in the gaseous diffusion process. The expression $P(I+AP)$ in Eq. 1-1 represents the modified pressure in the traditional ideal gas equation. The parameter A in the above expression represents the temperature-dependent van der Waals coefficient. Given values for the van der Waals coefficient at two temperatures, the following expression was derived that describes the nature of the van der Waals coefficient (atm⁻¹) as a function of UF₆ temperature (F).

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$$A = -2.02 \times 10^{-4} T + 0.0617 \text{ atm}^{-1}$$

Naturally, as the temperature of the gas increases, the gas behaves in a more ideal manner. For standard conditions in which the temperature is given as 59 F, the van der Waals coefficient is given as $0.049782 \text{ atm}^{-1}$, or 0.0034 psia^{-1} . Using the van der Waals coefficient at standard conditions, the above expression evaluates the modified van der Waals pressure as

$$P' = 14.7 \times (1 + (0.0034)(14.7)) = 14.75 \text{ psia.}$$

5.3 Calculation of the Monthly UF_6 and Uranium Release Estimate

Table 5.1-1 depicts a sample spreadsheet of the raw purge data for a three shift daily log from the Area 5 Foreman Logbook for the month of July 1975. The spreadsheets for the remaining months are in the Microsoft® Excel workbook RAWPURGE.XLS. The three data points for each day were averaged in order to provide daily averages of the purge rate (scfd) and Tops Conc (mol wt.% UF_6). For example, the daily average purge rate for July 1, 1975 would be

$$\frac{9500 + 11000 + 12500}{3} = 11000 \text{ scfd,}$$

and the daily average UF_6 concentration for July 1, 1975 would be

$$\frac{0.00001 + 0.00001 + 0.00001}{3} = 0.00001 \text{ mol wt. \% } \text{UF}_6.$$

Table 5.1-2 depicts a sample daily purge rate data spreadsheet for the month of July 1975. The spreadsheets for the remaining months are in the Microsoft® Excel workbook 95013R0.XLS. The purge flow is the sum of the purge rate in the purge unit. The daily molecular weighted UF_6 fraction in the purge gases is computed by multiplying the purge rate and the tops concentration. These daily rates are shown in the rightmost column in Table 5.1-2. For example, the per cent volume of UF_6 purged on July 1, 1975

$$(11000 \times 0.00001) + (0 \times 0) = 0.11 \text{ \%scfd.}$$

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Table 5.1-1: Raw Three Shift Purge Data Spreadsheet for July 1975

Purge Cell -- K-311-1						
Month -- July 1975						
12 am - 8 am			8 am - 4 pm		4 pm - 12 pm	
Day	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	Tops Conc
1	9500	0.00001	11000	0.00001	12500	0.00001
2	12000	0.00001	12500	0.00001	12500	0.00001
3	11500	0.00001	12000	0.00001	15000	0.00001
4	16000	0.00001	11500	0.00001	10000	0.00001
5	12500	0.00001	12500	0.00001	12500	0.00001
6	12500	0.00001	20000	0.00001	14000	0.00001
7	11500	0.00001	11500	0.00001	18000	0.00001
8	16000	0.00001	17000	0.00001	17000	0.00001
9	7000	0.00001	13000	0.00001	10500	0.00001
10	14000	0.00001	15000	0.00001	12000	0.00001
11	12000	0.00001	9500	0.00001	22000	0.00001
12	10000	0.00001	11500	0.00001	11000	0.00001
13	11000	0.00001	10500	0.00001	11000	0.00001
14	20000	0.00001	10500	0.00001	11000	0.00001
15	11000	0.00001	10500	0.00001	11000	0.00001
16	10500	0.00001	10500	0.00001	10000	0.00001
17	10000	0.00001	10000	0.00001	9000	0.00001
18	9000	0.00001	9000	0.00001	9500	0.00001
19	9500	0.00001	9000	0.00001	9500	0.00001
20	9500	0.00001	9500	0.00001	9500	0.00001
21	9500	0.00001	9500	0.00001	9000	0.00001
22	5000	0.00001	5000	0.00001	9000	0.00001
23	9000	0.00001	9000	0.00001	10500	0.00001
24	9000	0.00001	9000	0.00001	9000	0.00001
25	9000	0.00001	9000	0.00001	9000	0.00001
26	9000	0.00001	9000	0.00001	9000	0.00001
27	9000	0.00001	9000	0.00001	9000	0.00001
28	N/A	0.00001	9000	0.00001	9000	0.00001
29	9500	0.00001	9500	0.00001	9500	0.00001
30	9500	0.00001	9500	0.00001	9500	0.00001
31	9500	0.00001	9500	0.00001	9500	0.00001

The total volume of UF_6 purged for the month is the sum of these daily volumetric flows divided by 100. For July 1975, this computes to 0.034 ft^3 . The total volume of UF_6 at standard conditions is used in Eq. 1-1 to estimate the mass of UF_6 released from the purge unit for the month. The modified van der Waals pressure is substituted for the expression $P(1+AP)$. This calculation for July 1975 precedes as follows

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$$m = \frac{(14.75)(0.034)(453.6)}{(0.030488501)(519)} = 14.31 \text{ g UF}_6.$$

The uranium-only fraction of the UF_6 mass is calculated by multiplying by the ratio of the molecular weights of U and UF_6 computed in Section 5.1. For July 1975, the calculation precedes as

$$m = \frac{(14)(237.9442315)}{351.9346495} = 9.672 \text{ g U.}$$

5.4 Calculation of the Specific Activities

The "effective" specific activity of uranium is calculated using the expression in Eq. 1-2 and depends upon the enrichment of ^{235}U . Assuming an enrichment of 3.5% ^{235}U and the "effective" specific activity follows as

$$S = (0.4 + 0.38(3.5) + 0.0034(3.5)^2) \times 10^{-6} = 1.7717 \times 10^{-6} \text{ Ci/g.}$$

For July 1975, the "effective" activity of uranium is calculated by multiplying this value by the mass of uranium calculated in Section 5.3 and given as

$$A = (9.672)(1.7717 \times 10^{-6}) = 1.714 \times 10^{-5} \text{ Ci U.}$$

The specific activities of isotopic ^{234}U , ^{235}U , and ^{238}U are calculated using Eq 1-4. The decay constant, λ_i , for each isotope is given by Eq 5.4-1 as

$$\lambda_i = \frac{\ln 2}{T_{1/2}} \quad (5.4-1)$$

where $T_{1/2}$ is the half-life of the radioisotope.

The decay constant is typically expressed in units of s^{-1} and therefore requires conversion of the half-life to units of s. For the ^{234}U isotope, the decay constant is computed as

$$\lambda = \frac{\ln 2}{(2.46 \times 10^5)(365.25)(24)(3600)} = 8.92866 \times 10^{-14} \text{ s}^{-1}.$$

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The decay constants for ^{235}U and ^{238}U are similarly calculated. Using Eq 1-4 and converting units of Bq to units of Ci, the specific activity of isotopic ^{234}U is calculated as

$$S = \frac{(8.92866 \times 10^{-14})(6.022045 \times 10^{23})}{(234.040904)(3.7 \times 10^{10})} = 6.2092 \times 10^{-3} \text{ Ci/g.}$$

The specific activities of isotopic ^{235}U and ^{238}U are similarly calculated.

Table 5.1-2: Spreadsheet of Purge Rate Data for July 1975

Day of Month	311-1		None		Total	UF6 Purge
	Purge Rate	Tops Conc	Purge Rate	Tops Conc	Purge Rate	
1	11000	0.00001			11000	0.11
2	12333	0.00001			12333	0.123333333
3	12833	0.00001			12833	0.128333333
4	12500	0.00001			12500	0.125
5	12500	0.00001			12500	0.125
6	15500	0.00001			15500	0.155
7	13667	0.00001			13667	0.136666667
8	16667	0.00001			16667	0.166666667
9	10167	0.00001			10167	0.101666667
10	13667	0.00001			13667	0.136666667
11	14500	0.00001			14500	0.145
12	10833	0.00001			10833	0.108333333
13	10833	0.00001			10833	0.108333333
14	13833	0.00001			13833	0.138333333
15	10833	0.00001			10833	0.108333333
16	10333	0.00001			10333	0.103333333
17	9667	0.00001			9667	0.096666667
18	9167	0.00001			9167	0.091666667
19	9333	0.00001			9333	0.093333333
20	9500	0.00001			9500	0.095
21	9333	0.00001			9333	0.093333333
22	6333	0.00001			6333	0.063333333
23	9500	0.00001			9500	0.095
24	9000	0.00001			9000	0.09
25	9000	0.00001			9000	0.09
26	9000	0.00001			9000	0.09
27	9000	0.00001			9000	0.09
28	9000	0.00001			9000	0.09
29	9500	0.00001			9500	0.095
30	9500	0.00001			9500	0.095
31	9500	0.00001			9500	0.095
Total :	338333		0		338333	3.383333333
Average :	10914		0		10914	1.0000E-05
Volume UF6 :	0.034	ft^3				

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5.5 Calculation of the Isotopic Activity and Mass of ^{234}U , ^{235}U , and ^{238}U

An estimate of the contribution to the total "effective" activity of each uranium isotopic constituent is calculated according to Eq 5.5-1.

$$A_i = f_i S \quad (5.5-1)$$

where f_i is the fraction of total activity for each isotopic constituent from Fig 1-2, and S is the specific activity from Eq 1-2.

The fraction of total "effective" activity, f_i , for each uranium isotope is read from the graph in Fig 1-2 at the assumed 3.5% ^{235}U enrichment. The accompanying table at the right details the fractions used for each isotope in this calculation. For July 1975, the contribution to the total "effective" activity by ^{234}U is

i	f_i
^{238}U	0.183
^{235}U	0.043
^{234}U	0.774

$$A = (0.774)(1.714 \times 10^{-5}) = 1.326 \times 10^{-5} \text{ Ci } ^{234}\text{U}.$$

The contributions by ^{235}U and ^{238}U are calculated similarly. The estimate of the mass of each uranium isotopic constituent is calculated using equation Eq 1-3 and uses the theoretical specific activities for each uranium isotope computed in Section 5.4. For July 1975, the mass of ^{234}U is calculated as

$$m = \frac{1.326 \times 10^{-5}}{6.2092 \times 10^{-3}} = 2.136 \times 10^{-3} \text{ g } ^{234}\text{U}.$$

The mass of ^{235}U and ^{238}U are calculated similarly. The masses of the three isotopes were summed and compared with the total uranium mass calculated in Section 5.3 to ensure close agreement. The ratio of the ^{235}U mass to the total U mass was computed to ensure an approximate 3.5% enrichment level. The activity fractions in the table above represent the values resulting from several iterative refinements. The final fraction values from Fig 1-2 used in this calculation result in an overstatement of the total uranium mass by 0.05% over the estimate resulting from the van der Waals real gas equation. The uncertainties in the experimental data and the fitted equation in Fig 1-1 and in selecting values from the graph in Fig 1-2 contribute to the differences in the mass calculations. Also notable is the omission of the ^{234}U contribution to the U and UF_6 molecular weight calculations in Section 5.1 which impact the van der Waals real gas equation calculations in Section 5.3, the specific activity calculations in Section 5.4, and the mass calculations in Section 5.5.

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5.6 Discussion of the Results of the Calculation

This calculation estimates that between July 1975 and June 1976, about 142.1 g of slightly enriched uranium was vented to the atmosphere by the purge cascade. The monthly median during this one year period was about 11.5 g uranium with a standard deviation of about 2.7 g uranium. This release occurred daily and constituted a total of about 252 μCi with a monthly median of about 20 μCi and a standard deviation of about 4.7 μCi .

Gaseous diffusion causes a greater percent increase in ^{234}U than ^{235}U due to the better separation factor for ^{234}U . The half-life of ^{234}U is four and five orders of magnitude shorter than ^{235}U and ^{238}U and thus constitutes a higher percent of the overall activity. At 3.5% ^{235}U enrichment, about 77.4% of the total activity is due to the presence of $^{234}\text{UF}_6$ even though the weight percent of ^{234}U at this enrichment is only about 0.022%.

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7. ELECTRONIC FILES

The following files are included on the diskette in a self-extracting compressed format. The compressed file is named 95013R0.EXE.

<u>File Name</u>	<u>Description</u>
95013R0.DOC	This document (Microsoft® Word version 6.0a).
95013R0.XLS	Spreadsheet for the calculation of the ORGDP purge cascade uranium release estimates for July 1975 through June 1976 (Microsoft® Excel version 5.0a).
RAWPURGE.XLS	Spreadsheet for the raw purge rate data from the K-25 Area 5 Forman Logbooks (Microsoft® Excel version 5.0a).

Attachment 1

Purge Rates July 1975 - June 1976
(from the K-25 Area 5 Foreman Logbooks at K-1034-A Site Records)

MONTH JUL 1975MONTH AVG 1975422
0000
1695

DAY	CELL <u>K-311-1</u>						CELL <u>K-311-1</u>					
	SHIFT 1		SHIFT 2		SHIFT 3		SHIFT 1		SHIFT 2		SHIFT 3	
	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC
1	9500	00001	11000	00001	12500	00001	9500	00001	10500	00001	10000	00001
2	12000		12500		12500		10000		10000		10000	
3	11500		12000		15000		10000		10000		—	—
4	16000		11500		10000		10000		10000		9000	00001
5	12500		12500		12500		10500		11500		11000	
6	12500		20000		14000		11000		10500		11000	
7	11500		11500		18000		12500		10000		10000	
8	16000		17000		17000		10500		10000		10000	
9	7000		13000		10500		10000		10000		10500	
10	14000		15000		12000		11000		12000		10500	
11	12000		9500		22000		10000		11000		10500	
12	10000		11500		11000		6000		6000		10500	
13	11000		10500		11000		20000		20000		21000	
14	20000		10500		11000		10000		14000		11500	
15	11000		10500		11000		10000		12000		12000	↓
16	10500		10500		10000		8000		11000		—	—
17	10000		10000		9000		13000		11000		12000	00004
18	9000		9000		9500		12000		13000	↓	10000	00001
19	9500		9000		9500		12500		—	—	12500	
20	9500		9500		9500		12500		12500		11500	
21	9500		9500		9000		12000		12000		11500	
22	5000		5000		8000		12500		12500		16000	
23	9000		9000		10500		19500		11500		11500	
24	↓		↓		9000		13500		22000		11500	
25	↓		↓		↓		10500		10500		11000	
26	↓		↓		↓		10000		10500		10500	
27	↓		↓		↓		10500	↓	10000		10500	
28	—		↓		↓		—	—	20000		—	
29	9500		9500		9500		28000	00001	10500		10000	
30	↓		9500		↓		10500	11	10500		10500	
31	↓		9500	↓	↓	↓	10500	↓	10500	↓	10500	↓

MONTH SEPT 1976MONTH OCT 1976

Notes Taken by Jim Bennett 11/2/95

W
082
11/6/95CELL K-311-1CELL K-311-1

DAY	CELL <u>K-311-1</u>						CELL <u>K-311-1</u>					
	SHIFT 1		SHIFT 2		SHIFT 3		SHIFT 1		SHIFT 2		SHIFT 3	
	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC
1	10500	00001	10500	00001	10500	00001	10000	00001	11500	00001	10000	00001
2			10500		10000		10000		12000	00001	12000	00001
3			15000		10000		12000	—		OUT	12000	OUT
4			10500		10500			OUT			17000	
5	✓		10500								12000	
6	10000		10000								16000	
7	10500		10500								14000	↓
8							↓		↓		11500	00001
9							11500	↓	11500	↓	12500	
10							—	—	12000	00001	12500	
11							12500	00001	12000		12000	
12	↓		↓		↓		13500		10000			
13	12000		12000		11000		13500	↓	13000			
14	11000		11000		10500		—	—	18000			
15	10500		10500		10500		12500	00001	13000		↓	
16	15000		10500		10000		13000		13000		12500	
17	10500		10500		10000		12500		—			
18	—	—	10000		10500		12500		12500			
19	10000	00001			12000		13000		12500			
20	10000				10000				12500			
21	9500								13000			
22	10000								13000		↓	
23									14000		13000	
24									13000		13000	
25									13000		13000	
26							—	↓	13000		14000	
27					00001		↓	OUT	14000		10000	
28							15000	00001	13000	↓	13000	
29							13000			00002	13000	
30	↓	↓	↓	↓	↓	↓	13000	↓	↓	00001	12000	↓
31							12000	↓	↓	00001	13000	OUT

MONTH DEC 1975

MONTH NOV 1975

11/6/95

DAY	CELL 16.311-1						CELL 16.311-1					
	SHIFT 1		SHIFT 2		SHIFT 3		SHIFT 1		SHIFT 2		SHIFT 3	
	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC
1	14000	00001	14500	00001	14000	00001	13000	00001	12000	00001	13000	00001
2	14000		13500		13500		13000		13000		13000	00001
3	13500		13500		14500		13000		11000		11000	OUT
4	14000		14500		14000	↓	11000		11000		11000	OUT
5	14000		15000		—	—	11000		12000		11500	00001
6	13000		13000		13000	00001	11600		12000		11000	
7	13000		13000		12500		11000		11000		11000	
8	13000		13000		13000		11000		11000		11000	
9	13000		12500		13000		11000		11000		11000	
10	12000		12500		12000		11000		11000		11000	
11	13000		13000		13000		11000		11000		11000	
12	12500		13000		13000		11000		11000		11000	
13	12500		12500		13000		11000		11000		11000	
14	12500		12500		13000		10500		10500		11000	
15	13000		12500		12500		11000		10500		10500	
16	12000		12500		12500		11000		10500		11500	
17	12000		12500		12500		11000		11500		11000	
18	12500		12500	↓	12500		11000		11000		11000	
19	12000		—	↓	11500		11000		11000		11000	
20	11500		12000	00001	11500		12500		11000		11500	
21	11500		11500		11500		11500		11500		11500	
22	11500		12000		12000		12000		13000		11500	
23	12000		12500		12500		12500		13000		13000	
24	12500		12500		12500		12500		13500		13000	
25	12500		12500		12500		13000		15000		13000	
26	12500		12500		14000		13500		13500		13500	
27	12000		12000		12000		13500		13500		13500	
28	12000		12000	↓	12000		15000		15000		14000	
29	12000		12000	—	12000		14000		14000		14000	
30	12000		12500	00001	12000		14000		14000	↓	14000	↓
31	15000	↓	15000	00001	13000	↓						

MONTH JAN 1976

MONTH FEB 1976

4
082
116195

DAY	CELL K-311-1						CELL K-311-1					
	SHIFT 1		SHIFT 2		SHIFT 3		SHIFT 1		SHIFT 2		SHIFT 3	
	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC
1	12000	00001	12500	00001	12500	00001	16500	00001	16000	00001	17000	00001
2	12500		12500		12500		16500		16500		17000	
3	20000		12500		12500		16500		16000		16000	
4	13000		↓		↓		16000		16000		16000	
5	↓		↓		↓		16000		16000		14500	
6	↓		25000	—	13000		14500		14500		26000	
7	↓		11500	00001	"		14000		16000		14500	
8	14500		13500		14000		16000		14500		14500	
9	16000		18000		14500		16000		16000		16000	
10	14500		13500		14500		14000		14000		14000	
11	13000		24000		13000		13500		13000		13000	
12	13000		13000		13000		13500		14000		15000	
13	13000		↓		↓		14500		14500		14500	
14	↓		↓		↓		14500		16000		15500	
15	↓		↓		12000		16000		14000		14000	
16	↓		↓		"		14000		14000		14500	
17	12000		15000		15000		14000		14000		14000	
18	16000		15500		15500		14000		13500		14000	
19	"		"	"	"		14000		13500		14000	
20	15500		15000		15000		13500		13500		13500	
21	15000		↓		15500		13000		13500		13500	
22	15500		↓		16000		14000		13500		13500	↓
23	↓		↓		15000		11500		14500	↓	16000	—
24	↓		↓		14500		13500		14000	—	12500	00001
25	14000		14000		14000		11000		13000	00001	13000	
26	"		↓		15000		12000		12500		16500	↓
27	15000		↓		13500		12500	↓	—	00001	19500	00001
28	14000		↓		14000		13000	00001	14000	00001	14000	00001
29	↓		↓		"		14000	00001	13500	00001	13500	00001
30	↓		↓		17000		"					
31	↓		↓		16000	↓						

MONTH MARCH 1976MONTH APRIL 19764
as
11/6/76

DAY	CELL <u>K-311-1</u>						CELL <u>K-311-1</u>					
	SHIFT 1		SHIFT 2		SHIFT 3		SHIFT 1		SHIFT 2		SHIFT 3	
	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC
1	13000	00001	13500	00001	13000	00001	13500	00001	13000	00001	13500	00001
2	30000		11500		13000		13500		13500		13500	
3	13000		13000		13000		13500		13500		13000	
4	13000		13000		13000		13500		13000		13500	
5	13000		12000		14500		13500		13000		13000	
6	15000		15000		14500		13500		13500		13500	
7	14500		14500		14000	↓	13500		13500		13500	
8	13500		11500		—	00001	13500		13500		13500	
9	15000		15000	↓	15000		13500		13500		14000	
10	14500		—	—	15000		13500		13500		13500	
11	15000		11500		12000		13500		13500		13500	
12	15000		15000		14500		13500		17000		15500	
13	12500		13000		13000		15000		13500		15000	
14	14000		14500		14000		15000		13500		13500	
15	14000		14000		14000		13500		13500		13500	
16	14000		14000		13500		19000	↓	12000		13000	
17	15000		10500		17000		13000	—	13000		13500	
18	10500		11000		14000		13000	00001	13000		13500	
19	15000		14000	↓	13500		13500		15000		13500	↓
20	14000		13500		13500		13500		13000		12500	—
21	13500		13000		13500		13000		14000	↓	18000	00001
22	12000		13000		14000		13000	↓	—	—	15000	—
23	13500		14000		14000		—	—	13000	00001	13000	—
24	14000		14000		14000		13800	00001	15000	—	13000	—
25	14000		14000		13000		12500		13500	—	12600	—
26	14000		14000		12000		14000		13500	—	13500	—
27	14000		14000		13000		14500		14000	—	13500	—
28	14000	↓	14000		14000		19500		—	—	—	—
29	13500	—	14000		12500		80000	↓	40000	—	18500	00001
30	13500	00001	12500		12000		22000	↓	14000	00001	22000	—
31	13000	00001	12500	↓	12500	↓						

MONTH MAY 1976

MONTH JUNE 1976

11/22/76
11/6/76

CELL K-311-1

CELL KC-311-1

DAY	SHIFT 1		SHIFT 2		SHIFT 3		SHIFT 1		SHIFT 2		SHIFT 3	
	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC	PURGE	TC
1	20000	—	19000	—	22000	—	14500	00001	14000	00001	15000	00001
2	20000	—	—	—	14000	00001	14500		14500		15000	
3	17500	00001	20000	00001	23000	00001	14500		14500		15000	
4	15500		17500		19000		16000		15500		10500	
5	15000		17000		20000		10500		16500		10500	
6	14500		16000		15000		10500		15300		11500	
7	14500		15000		15500		15000		15500		15000	
8	17000		15500		25000		15300		45000		15000	
9	20000		15300		15500		16000		14000		15000	
10	16000		15500		15500		14000		14300		14500	
11	15500		15000		15300		14500		14500		14500	
12	16000		15000		17000		14000		14500		14500	
13	15500		18000		15000		15000		14500		14500	
14	15000		15000		15000		14500		15500		14500	
15	15000		15000		15000		14500		15000		15300	
16	15000		15000 16000		18000		15000		15000		15000	
17	17000 16000		10000		16000 15500		14500		15000		14500	
18	15500		15500 15000		18000		14500		14500		14500	
19	15500 16000		18000		17000 15500		15000		15000		25000	
20	15500 16000		17000		14500		15000		15000		14500	
21	15500		19500		15500		15000		15000		14500	
22	14500 14000		17000 16500		14500 14000		15000		15000		14500	—
23	15000		15500 15000		18000		14500		15000		—	—
24	15000		15000		19000		14500		14500		15000	00001
25	15000		15000		15000		15000		15000		15000	
26	15000		16000		14500 15000		26500		36000		15000	
27	14500 15000		20000 19000		15000 14500		26500		28000		19000	
28	14500 15000		20000 19000		15000 14500		12000		16000		17000	
29	14500		14500 14000		15000		16000		16000		—	—
30	15000		15000		15000		16000		17000		19000	
31	—		15000		15000							

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CALC NO **SRA-96-012** **REV 0**

Project/Task ChemRisk/TDH Oak Ridge Dose Reconstruction

Prepared by R. E. Burmeister

Date 11/1/96

Checked by/Date

J. J. Shonka

Title The Atmospheric Master Release List and Atmospheric Source Term for K-25

(signed original on file)

SHONKA RESEARCH ASSOCIATES, INC.

Calculation Control Sheet

Calculation number: SRA-96-012 REV. 0

Title: The Atmospheric Master Release List and Atmospheric Source Term for K-25

Reason for calculation/revision: New calculation

Client: ChemRisk/TDH

Project: Oak Ridge Dose Reconstruction

Project/Task Number: Task 6

Prepared by: Regan E. Burmeister
Regan E. Burmeister (signed original on file)

Date: 11/1/96

Independent Technical Review by:

Joseph J. Shonka (signed original on file)

Date: 11/1/96

Quality Assurance Review by: Deliah B. Sharke

Date: 11/1/96

Deborah B. Shonka (signed original on file)

☐ This calculation has been voided or superseded by

(calculation number)

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CALC NO SRA-96-012 REV 0Project/Task ChemRisk/TDH Oak Ridge Dose ReconstructionPrepared by R. E. BurmeisterDate 11/1/96Checked by/Date J. J. ShonkaTitle The Atmospheric Master Release List and Atmospheric Source Term for K-25

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Review Method Sheet

The undersigned has reviewed this calculation in accordance with the method(s) indicated below.

1. Computer Aided Calculation	
a	Review to determine that the computer program(s) has been validated and documented, is suitable to the problem being analyzed, and that the calculation contains all necessary information for reconstruction at a later date.
b	Review to determine that the input data as specified for program execution is consistent with the design input, correctly defines the problem for the computer algorithm and is sufficiently accurate to produce results within any numerical limitations of the program.
c	Review to verify that the results obtained from the program are correct and within stated assumptions and limitations of the program and are consistent with the input.
d	Review validation documentation for temporary changes to listed, or developmental, or unique single application programs, to assure that the methods used adequately validate the program for the intended application.
e	Review of code input only, since the computer program has sufficient history of use at Shonka Research Associates, Inc. in similar calculations.
f	Review arithmetic necessary to prepare code input data.
g	Other:
2. Hand Prepared Calculations	
a	Detailed review of the original calculations.
b	Review by an alternate, simplified, or approximate method of calculation.
c	Review of a representative sample of repetitive calculations.
d	Review of the calculation against a similar calculation previously performed.
e	Other:
3. Revisions	
a	Editorial changes only
b	Elimination of unapproved input data without altering calculated results.
c	Other:
4. Other	

Reviewed by

Joseph J. Shonka (signed original on file)

Date:

11/1/96

SHONKA RESEARCH ASSOCIATES, INC.

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CALC NO SRA-96-012 REV 0

Project/Task ChemRisk/TDH Oak Ridge Dose Reconstruction

Prepared by R. E. BurmeisterDate 11/1/96Checked by/Date J. J. Shonka

Title The Atmospheric Master Release List and Atmospheric Source Term for K-25

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ABSTRACT

This calculation presents the ORHS-II Atmospheric Master Release List for the Oak Ridge Gaseous Diffusion Plant. The list is an Excel spreadsheet with many columns and sheets that detail the construction of the uranium release history from 1945 to 1995. All of the data used in the list are given references. The formulas for certain columns are explained. In particular, the use of the results of previous calculations to fill chronological gaps in the release history are detailed.

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Project/Task ChemRisk/TDH Oak Ridge Dose Reconstruction

Prepared by R. E. Burmeister

Date 11/1/96

Checked by/Date

J. J. Shonka

Title The Atmospheric Master Release List and Atmospheric Source Term for K-25

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Appendix A Copies of two personal communications
Task 6: Review of Release Fraction Literature
1986-1995 K-25 Uranium Airborne Releases

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Project/Task ChemRisk/TDH Oak Ridge Dose Reconstruction

Prepared by R. E. Burmeister

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1. INTRODUCTION

Task 6 of the Oak Ridge Dose Reconstruction focuses on the evaluation of the quality of historical airborne and waterborne effluent monitoring data and the determination of the potential significance of unmonitored emissions. Uranium played an important role throughout historical operations on the Oak Ridge Reservation (ORR) and is known to have been released to the environment through air and water. The two largest uses of uranium on the Reservation were the enrichment processes of the ^{235}U isotope by electromagnetic separation at the Y-12 facility and gaseous diffusion at the K-25 facility.

This calculation focused on atmospheric uranium releases from the gaseous diffusion process at the K-25 site. The K-25 site was comprised of the five different cascade complexes, K-25, K-27, K-29, K-31, and K-33, as well as many buildings that supported the gaseous diffusion process. Included as part of the K-25 site for this calculation was the S-50 liquid thermal diffusion plant (SRA-96-011). Many releases were accidents due to equipment failures or personnel mistakes. Other releases were scheduled and deliberate, such as the releases from the purge cascade. Releases through other pathways that did not include the atmosphere were collected and documented during the search for material release events. These releases were not included in this calculation.

As much information as possible about each release was gathered. Releases were typified by their date of occurrence and amount of release as well as other information that allowed the releases to be classed according to release pathway or location of release. It was known that certain gaps in the historical releases from certain buildings existed; release data was unavailable or did not exist but these buildings were operational for known time frames. The results of two previous calculations, SRA-96-009 and SRA-96-010, were used to fill these gaps.

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Prepared by R. E. BurmeisterDate 11/1/96Checked by/Date J. J. Shonka

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2. SUMMARY OF RESULTS

Table 2.1 and Table 2.2 give the chronological atmospheric uranium release history for the Oak Ridge Gaseous Diffusion Plant (ORGDP) for the 50% and 95% certainty values, respectively. Care should be taken in using these values.

The two certainty values were used to bound the release values. These certainty values were the results of statistical fits to some of the release data (SRA-96-009 and SRA-96-010). In all other cases, when releases were not fit, no uncertainty was asserted. Thus, the actual uncertainty in the release results was larger than has been expressed in this calculation. Only releases whose pathway was evaluated as being released to the atmosphere were included here.

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CALC NO SRA-96-012 REV 0

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**Table 2.1 Uranium Release History for 50% Certainty
Atmospheric Pathways Yearly Release**

Date	Uranium (kg)	Uranium (Ci)	U-235 (kg)	U-238 (kg)	U-235 (Ci)	U-238 (Ci)	U-234 (Ci)	Uranium Cumulative (kg)	Uranium Cumulative (Ci)
1944	412.5	0.27716	2.9	409.6	0.00634	0.13766	0.13316	412.5	0.27716
1945	1246.7	0.83816	8.9	1237.8	0.01918	0.41605	0.40292	1659.2	1.11532
1946	115.8	0.19671	3.4	112.4	0.00729	0.03778	0.15163	1775.0	1.31202
1947	115.5	0.21226	3.5	112.0	0.00754	0.03764	0.16708	1890.5	1.52428
1948	119.9	0.20789	3.4	116.4	0.00732	0.03912	0.16145	2010.4	1.73217
1949	192.9	0.24933	3.7	189.1	0.00802	0.06356	0.17774	2203.2	1.98150
1950	250.8	0.29517	4.3	246.1	0.00930	0.08272	0.20315	2454.0	2.27667
1951	707.4	0.45399	7.3	700.0	0.01587	0.23528	0.20284	3161.4	2.73066
1952	1211.7	0.83800	8.6	1203.1	0.01854	0.40438	0.41508	4373.1	3.56866
1953	1307.0	1.20813	17.2	1289.8	0.03718	0.43352	0.73743	5680.1	4.77679
1954	459.1	0.98574	15.4	443.7	0.03325	0.14912	0.80337	6139.1	5.76253
1955	482.7	0.40827	6.1	476.6	0.01313	0.16020	0.23493	6621.9	6.17079
1956	397.1	0.45520	6.5	390.7	0.01396	0.13132	0.30993	7019.0	6.62599
1957	442.8	0.50339	7.1	435.7	0.01540	0.14645	0.34154	7461.8	7.12938
1958	2706.8	2.19421	27.7	2678.6	0.05992	0.90032	1.23397	10168.6	9.32359
1959	675.8	0.88153	12.7	661.9	0.02735	0.22248	0.63171	10844.4	10.20513
1960	1189.6	0.89979	10.7	1178.9	0.02307	0.39624	0.48048	12033.9	11.10491
1961	896.9	0.75221	8.9	888.0	0.01932	0.29846	0.43443	12930.8	11.85712
1962	163.5	0.34328	6.0	157.5	0.01288	0.05295	0.27744	13094.4	12.20040
1963	1003.3	8.49670	182.7	820.6	0.39483	0.27582	7.82605	14097.7	20.69711
1964	23.6	0.09661	1.8	21.8	0.00379	0.00733	0.08549	14121.3	20.79372
1965	456.5	0.86642	16.7	439.8	0.03609	0.14782	0.68251	14577.8	21.66014
1966	19.4	0.09083	1.7	17.7	0.00361	0.00595	0.08128	14597.1	21.75098
1967	18.8	0.09004	1.7	17.1	0.00358	0.00576	0.08070	14615.9	21.84102
1968	20.6	0.09107	1.7	18.9	0.00360	0.00637	0.08111	14636.6	21.93209
1969	28.8	0.09718	1.7	27.1	0.00376	0.00910	0.08432	14665.4	22.02927
1970	24.8	0.09672	1.8	23.0	0.00382	0.00774	0.08516	14690.2	22.12599
1971	70.1	0.18272	3.5	66.4	0.00753	0.02232	0.15287	14760.2	22.30870
1972	44.0	0.11490	2.0	42.0	0.00440	0.01411	0.09639	14804.3	22.42360
1973	284.4	0.44241	9.5	274.9	0.02046	0.09240	0.32955	15088.6	22.86601
1974	621.9	1.45982	31.5	590.4	0.06798	0.19843	1.19341	15710.5	24.32583
1975	370.9	0.76303	16.6	354.3	0.03587	0.11907	0.60810	16081.4	25.08886
1976	114.4	0.25340	5.9	110.7	0.01275	0.03722	0.20343	16195.8	25.34226
1977	36.5	0.12694	2.4	34.0	0.00521	0.01144	0.11029	16232.3	25.46920
1978	28.1	0.10481	1.9	26.1	0.00418	0.00879	0.09183	16260.4	25.57400
1979	33.6	0.11000	2.4	31.2	0.00518	0.01050	0.09432	16294.0	25.68400
1980	121.7	0.20170	4.8	116.9	0.01037	0.03929	0.15204	16415.7	25.88571
1981	68.5	0.13000	3.5	65.0	0.00761	0.02185	0.10054	16484.2	26.01571
1982	73.7	0.11000	3.2	70.5	0.00690	0.02369	0.07942	16557.9	26.12571
1983	19.5	0.09116	1.7	17.8	0.00363	0.00598	0.08155	16577.4	26.21687
1984	19.3	0.09068	1.7	17.5	0.00361	0.00588	0.08119	16596.6	26.30755
1985	20.0	0.09059	1.7	17.4	0.00360	0.00586	0.08113	16616.6	26.39814
1986	0.2	0.00100	0.0	0.2	0.00005	0.00006	0.00089	16616.8	26.39914
1987	0.4	0.00030	0.0	0.4	0.00001	0.00013	0.00016	16617.2	26.39944
1988	1.7	0.00110	0.0	1.7	0.00002	0.00057	0.00051	16618.9	26.40054
1989	1.1	0.00040	0.0	1.1	0.00000	0.00037	0.00003	16620.0	26.40094
1990	2.0	0.00110	0.0	2.0	0.00002	0.00067	0.00041	16622.0	26.40204
1991	40.2	0.02400	0.2	40.0	0.00045	0.01345	0.01010	16662.2	26.42604
1992	112.4	0.06400	0.5	111.9	0.00108	0.03761	0.02531	16774.6	26.49004
1993	12.0	0.01000	0.1	11.9	0.00029	0.00399	0.00572	16786.6	26.50004
1994	10.0	0.00800	0.1	9.9	0.00023	0.00333	0.00445	16796.6	26.50804
1995	16.2	0.00670	0.0	16.2	0.00001	0.00545	0.00123	16812.9	26.51474
Totals	16812.9	26.51474	467.1	16344.4	1.00936	5.49360	20.01179		

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CALC NO SRA-96-012 REV 0

Project/Task ChemRisk/TDH Oak Ridge Dose Reconstruction

Prepared by R. E. Burmeister

Date 11/1/96

Checked by/Date

J. J. Shanka

Title The Atmospheric Master Release List and Atmospheric Source Term for K-25

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Table 2.2 Uranium Release History at 95% Certainty
Atmospheric Pathways Yearly Release

Date	Uranium (kg)	Uranium (Ci)	U-235 (kg)	U-238 (kg)	U-235 (Ci)	U-238 (Ci)	U-234 (Ci)	Uranium Cumulative (kg)	Uranium Cumulative (Ci)
1944	1287.5	0.86507	9.2	1278.3	0.01978	0.42967	0.41562	1287.5	0.86507
1945	3871.7	2.60189	27.5	3844.2	0.05952	1.29208	1.25029	5159.2	3.46696
1946	137.9	0.37671	3.9	134.0	0.00836	0.04504	0.32330	5297.1	3.84367
1947	138.4	0.44751	4.8	133.6	0.01032	0.04491	0.39228	5435.5	4.29118
1948	142.9	0.44313	4.7	138.0	0.01009	0.04639	0.38665	5578.3	4.73431
1949	215.8	0.48457	5.0	210.8	0.01080	0.07084	0.40294	5794.1	5.21889
1950	273.7	0.53041	5.6	267.7	0.01208	0.08999	0.42835	6067.8	5.74930
1951	882.6	0.72924	9.7	872.9	0.02092	0.29339	0.41493	6950.4	6.47854
1952	1212.5	0.89325	9.4	1203.2	0.02024	0.40440	0.46861	8162.9	7.37179
1953	1287.2	1.48746	18.9	1268.3	0.04076	0.42630	1.02041	9450.1	8.85925
1954	637.0	1.45242	19.7	617.3	0.04262	0.20748	1.20233	10087.1	10.31167
1955	597.2	0.85827	10.0	587.2	0.02155	0.19735	0.63936	10684.3	11.16993
1956	458.3	1.12045	11.1	447.2	0.02402	0.15032	0.94611	11142.6	12.29038
1957	504.0	1.16864	11.8	492.3	0.02546	0.16545	0.97772	11646.7	13.45902
1958	2702.0	2.78840	31.2	2670.3	0.06748	0.89754	1.82338	14348.7	16.24742
1959	737.0	1.54678	17.3	718.5	0.03741	0.24148	1.26789	15085.6	17.79420
1960	1211.8	1.08781	11.3	1200.5	0.02439	0.40350	0.65991	16297.4	18.88201
1961	919.0	0.93221	9.4	909.6	0.02039	0.30572	0.60610	17216.4	19.81422
1962	185.8	0.53130	6.6	179.1	0.01420	0.06021	0.45688	17402.2	20.34551
1963	1002.6	3.39361	68.5	934.1	0.14807	0.31397	2.93157	18404.8	23.73912
1964	61.9	0.52662	5.1	56.7	0.01108	0.01907	0.49647	18466.6	24.26574
1965	456.5	0.98499	12.8	443.7	0.02770	0.14912	0.80816	18923.1	25.25073
1966	57.6	0.52084	5.0	52.6	0.01090	0.01768	0.49226	18980.8	25.77157
1967	57.1	0.52005	5.0	52.1	0.01087	0.01750	0.49169	19037.8	26.29162
1968	58.9	0.52108	5.0	53.9	0.01088	0.01810	0.49209	19096.7	26.81270
1969	67.1	0.52718	5.1	62.0	0.01104	0.02083	0.49531	19163.8	27.33988
1970	63.1	0.52672	5.1	57.9	0.01111	0.01947	0.49615	19226.9	27.86660
1971	108.4	0.61272	6.9	101.3	0.01481	0.03405	0.56385	19335.2	28.47932
1972	82.3	0.54491	5.4	76.9	0.01169	0.02585	0.50737	19417.5	29.02423
1973	284.3	0.87241	11.7	272.6	0.02531	0.09164	0.75547	19701.9	29.89665
1974	621.9	1.88983	33.4	588.4	0.07221	0.19776	1.61985	20323.8	31.78647
1975	370.9	1.19304	18.7	352.2	0.04036	0.11836	1.03432	20694.6	32.97951
1976	114.4	0.68340	7.5	109.1	0.01630	0.03667	0.63043	20809.0	33.66291
1977	74.7	0.55694	5.8	68.9	0.01249	0.02317	0.52127	20883.8	34.21985
1978	66.4	0.53481	5.3	61.1	0.01147	0.02052	0.50282	20950.1	34.75466
1979	59.8	0.52658	5.2	54.7	0.01116	0.01837	0.49705	21010.0	35.28124
1980	121.7	0.63170	7.0	114.7	0.01503	0.03856	0.57811	21131.7	35.91294
1981	68.5	0.56001	5.4	63.1	0.01175	0.02120	0.52705	21200.2	36.47295
1982	73.7	0.54001	5.5	68.1	0.01187	0.02291	0.50523	21273.8	37.01295
1983	57.7	0.52117	5.0	52.7	0.01091	0.01771	0.49254	21331.6	37.53412
1984	57.5	0.52069	5.0	52.4	0.01089	0.01761	0.49218	21389.1	38.05481
1985	58.2	0.52060	5.0	52.3	0.01089	0.01759	0.49212	21447.3	38.57541
1986	0.2	0.00100	0.0	0.2	0.00005	0.00006	0.00089	21447.5	38.57641
1987	0.4	0.00030	0.0	0.4	0.00001	0.00013	0.00016	21447.9	38.57671
1988	1.7	0.00110	0.0	1.7	0.00002	0.00057	0.00051	21449.7	38.57781
1989	1.1	0.00040	0.0	1.1	0.00000	0.00037	0.00003	21450.8	38.57821
1990	2.0	0.00110	0.0	2.0	0.00002	0.00067	0.00041	21452.8	38.57931
1991	40.2	0.02400	0.2	40.0	0.00045	0.01345	0.01010	21493.0	38.60331
1992	112.4	0.06400	0.5	111.9	0.00108	0.03761	0.02531	21605.4	38.66731
1993	12.0	0.01000	0.1	11.9	0.00029	0.00399	0.00572	21617.4	38.67731
1994	10.0	0.00800	0.1	9.9	0.00023	0.00333	0.00445	21627.4	38.68531
1995	16.2	0.00670	0.0	16.2	0.00001	0.00545	0.00123	21643.6	38.69201
Totals	21643.6	38.69201	472.6	21169.6	1.02136	7.11542	30.55523		

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3. METHODS

An Excel spreadsheet was developed that primarily functioned as a database of uranium releases from the ORGDP. Documents were retrieved from record centers at the Oak Ridge Reservation (ORR). The documents were mainly accountability records that indicated when, where, why, what and how much material containing uranium was released. This information was entered into the database.

Interviews conducted with active and retired personnel help to confirm the type and scope of activities that occurred during their employment (Bennett 1995A)(Bennett 1995B)(Bennett 1995C)(Buddenbaum 1995)(Burmeister 1996)(Shonka 1995). Using this information and other historical documents, it could be determined whether or not the database had a complete release history for a particular ORGDP building. For those high priority buildings that did not have a complete history, reasonable estimates of releases were made using probability distributions (SRA-96-009, -010, -011). These estimates were added to the database for the particular time period during which they were applicable.

An annual release amount in kilograms and curies of uranium was determined by summing all release amounts for each year of operation from 1944 to 1995. These are the release amounts given in Tables 2.1 and 2.2.

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4. ASSUMPTIONS

The following assumptions were made in this calculation:

- 1) The Environmental Monitoring Reports for the Oak Ridge facilities reported curies released to the atmosphere. It was assumed that these releases were due entirely to the ORGDP (See *Environment* in Section 5.) This has overstated the releases in the post 1980 time frame.
- 2) It was assumed that the building release fraction for all buildings was unity; i.e., all releases inside buildings that had a pathway to the atmosphere were assumed to transport 100% to the atmosphere. (Appendix A, JJS.048*).
- 3) No corrections were made for sample line loss. Losses estimated from stack sampling may be significantly understated, perhaps by as much as a factor of 4 or more.
- 4) The trapping efficiency for the purge cascade releases was assumed to 85%; i.e. 15% of material was released to the atmosphere.(SRA-96-009).
- 5) Natural enrichment of 0.711% was assumed for the environmental release data of 1989. Release data were inconsistent to provide a proper yearly enrichment level.

* Private communication titled, "Task 6: Review of Release Fraction Literature". Copies to Tom Widner and Jack Buddenbaum. 8/22/96.

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5. CALCULATION

This section details how the Atmospheric Master Release List (the List) spreadsheets were constructed. The reader should refer to the electronic copy on the enclosed disk. The List has twelve worksheets titled as one scrolls left to right as follows: 1) *Atmospheric Release*, 2) *Atm. Yearly Release*, 3) *Cylinder Fire Test*, 4) *Cascade Fit (ESA)*, 5) *K-1131, K-1401, K-1420 Fits (ESA)*, 6) *Environment*, 7) *Environment 2*, 8) *10% Diff*, 9) *S-50*, 10) *Uranium*, 11) *New Data 10-31-96*, and 12) *New Data 8-29-96*. The descriptions of these sheets given below apply to both the 50% and 95% certainty fit values that were determined in the previous calculations. There are actually two spreadsheets; one for 50% certainty and one for 95% certainty. The release data are the same in the two spreadsheets; only the fit values change between the spreadsheets.

The worksheet *Atmospheric Release* contains a chronological listing of releases of uranium for the ORGDP. Releases are classified on their location, amounts of uranium, U-238, and U-235, weight percents U-235, and release pathways. Some descriptive notes and references are given for each release. This worksheet was assembled primarily from accountability records that were retrieved from records centers on the ORR. Releases were assessed into several pathways. The ESA pathway was used to describe releases from Equipment to Stacks or vents and thus to the Atmosphere. Other pathway categories were described in SRA-96-010.

The worksheet *Atm. Yearly Release* gives the total yearly release amounts for 1945 to 1995 in grams and curies of uranium, U-238, and U-235; curies of U-234 are also given. Cumulative totals are also given along with plots of the release amounts. Entries for a particular year are the sums of many terms. The releases listed in *Atmospheric Release* were summed for each individual year. To these sums were added contributions from *Cascade Fit (ESA)*, *K-1131, K-1401, K-1420 Fits (ESA)*, *Environment*, *Environment 2*, *10% Diff*, *S-50*, *New Data 10-31-96*, and *New Data 8-29-96*. These contributions fill gaps in the release data.

Jennifer K. Lamb

10/4/65 10 lbs.
 10/5/65 55 lbs.
 10/7/65 248.9
 10/14/65 53.05
 10/29/65 245.00

mates of releases that were used to fill in
 ars that data was unavailable or did not
 and *K-1131, K-1401, K-1420 Fits (ESA)*,
 ease history for the purge cascade and the
 31-96 contains releases that were
 SRA) on October 31, 1996. *New Data 8-*
 at Shonka Research Associates (SRA)
 work in assembling the List had already

urge cascade for all years of operation.
 1 purge cascade data existed. A null

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value implies that nothing more needed to be added to the yearly release amounts. The assumption of a 15% release fraction, per Assumption 4), was applied here.

K-1131, K-1401, K-1420 Fits (ESA) gives the fits to the K-1131, K-1401, and K-1420 buildings for the ESA pathway. The fitted values are shown along with factors that represent fractions of years. Some of the release data for these buildings had only a few months of releases for particular years. To fill in the rest of the year, the fractions multiplied the fit values. The results were added to the yearly release amounts. No information was found that indicated that the buildings' operations were any different or were shut down for those missing fractions of years. The release data were only unavailable or did not exist.

The next sheet, *Cylinder Fire Test*, records the releases of UF_6 that occurred in October 1965 as a part of UF_6 cylinder test and development. These releases occurred at the ORGDP Rifle Range and were regarded separately from gaseous diffusion process releases. These releases were added to yearly totals only after releases from *Environment* and *10% Diff*.

The sheet *Environment* gives the atmospheric discharges in curies of uranium for the years 1973 to 1982. The discharges were taken from Environmental Monitoring Reports for the Oak Ridge Facilities for the years 1973 to 1982 inclusive (US AEC 1973)(US ERDA 1974-1976)(US DOE 1977-1982). These reports provided all uranium released from all Oak Ridge Facilities. It was conservatively assumed that all releases were from K-25 operations since there was no way to separate the contributions from all facilities. The curies of uranium were converted into curies and grams of U-238 and U-235. These amounts were then compared to the amounts that had been determined up to this point. Where the difference between the environmental amount and the amount to this point was positive for a particular year, it was assumed that information was missing from the release history and the difference was added to that year's amount. This practice overstated the releases from K-25. The additions occurred on the sheet *Atm. Yearly Release*.

In *Environment* it was necessary to have an enrichment level in order to convert curies to grams. The environmental reports did not give any information regarding enrichment levels. For those years that had a positive difference, the release data was examined to determine the enrichment level. Two methods were used to get an enrichment level indicative of those years. In the first method, the release data for those years were examined to get an average enrichment level; in each year, each data point's enrichment level was summed to a total and then divided by the number of data points. In the second method, each data point's U-235 mass and uranium mass were summed to totals, and then the total U-235 mass was divided by the total uranium mass to get an enrichment level indicative of each year. These two methods are identified in *Environment*. The actual calculation of these enrichments occurred in the spreadsheet ENVIRO.XLS to which *Environment* contained a data link to these enrichments. The second

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method was chosen because the ratio of masses was the correct definition of a year's enrichment level. In this manner, yearly release data was reconciled with the environmental reports as far as Assumption 1) was concerned (all ORR site releases were due to K-25).

The sheet *Environment 2* gives environmental data from 1986 to 1995. The sheet contains the original environmental release data for K-25. The release data were kilograms and curies of uranium for the ten years 1986 through 1995. To get a breakdown of kilograms and curies for the nuclides U-238, U-235, and U-234, it was necessary to have the enrichment level. This was calculated by using an expression of alpha specific activity as a function of enrichment (EGG-2530). The resulting enrichment level was an expression of the yearly enrichment, but it was recognized that the values were not average enrichments since no data ^{were} available to assert average enrichments. One particular year, 1989, had a calculated enrichment that was negative. This implied that the release data for that year were inconsistent. Since only a small amount of uranium was reported that year, namely 1.11 kg, a natural enrichment of 0.711% was assumed. For such a small release amount, the assumption had a negligible effect on the site cumulative release, but the assumption was the determining factor for that year since the environmental release was the only datum for the year of 1989.

There was another inconsistency in the data for 1992. One reference* reported the release for 1992 as 112.39 kg of uranium at 0.0640 Curies of activity (Appendix A - Buddenbaum Memo). The environmental release report for 1995 reported five years of data and gave the 1992 release amounts as 14.49 kg of uranium at 0.0100 Curies of activity (ENVN-95). The larger values were used in this calculation in order to conservatively state the release for 1992.

The sheet *New Data 10-31-96* contains release data received and reviewed at SRA after primary work had already been completed in assembling yearly releases. No impact of this data was found on any of the prior work. This was mainly due to the releases having release amounts of only a few tens of grams of uranium. The data were added to the yearly releases.

The sheet *New Data 8-29-96* contains release data received at SRA after primary work had already been completed in assembling the yearly releases. This data was reviewed to see if it impacted any of the fitting work accomplished for the purge cascade, K-1131, K-1420, or K-1401. It was determined that there was no impact, and that the data could be added to yearly releases without modifying the previous work.

The yearly release totals to this point were next compared to the reported yearly release amounts from K/HS-95. For those years where the reported value was 10% or more greater than the determined value, the difference was added to the determined value. It was argued that for those

* Private communication titled, "1986-1995 K-25 Uranium Airborne Releases," from Jack Buddenbaum to Joseph Shonka with copies to Tom Widner and Jennifer Cockroft. 10/23/96.

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years, K/HS-95 had valid but unavailable data that had not yet been retrieved, and to account for such valid data, the differences were added. This analysis occurred on the sheet *10% Diff*.

The sheet *S-50* contains the releases estimates for the S-50 liquid thermal diffusion plant (SRA-96-011). S-50 operated from September 1944 to September 1945. It produced only low enriched uranium, never exceeding 1% enrichment in product.

The sheet *Uranium* gives physical data for the element uranium and its isotopes. This data was used as needed to calculate grams and curies.

Once all the above had been accomplished, the fit values were replaced with their 95% certainty values determined in the previous calculations. This resulted in two master spreadsheets called ATM10A.XLS and ATM10B.XLS which are included in the disks. ATM10A.XLS contains the 50% certainty work; ATM10B.XLS contains the 95% certainty work.

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6. REFERENCES

Bennett 1995A	Interview Notes: Jack Bailey. 10/10/95. Inmagic # 2857
Bennett 1995B	Interview Notes: Jim Rogers. 10/24/95. Inmagic # 2532
Bennett 1995C	Interview Notes: Joe Dykstra. 10/12/95. Inmagic # 2858
Buddenbaum 1995	Interview Notes: Dave Stoddard. 10/11/95. Inmagic # 2533
Burmeister 1996	Interview Notes: Jim Rogers. 9/10/96. Inmagic # 2927
EGG-2530	Department of Energy. Health Physics Manual of Good Practices for Uranium Facilities. EGG-2530. Idaho National Engineering Laboratory. 1988. p. 2-7
ENVN-95	Oak Ridge Reservation Annual Site Environmental Report for 1995. Chapter 3. Table 4.3, Fig. 4.7 and Fig. 4.8. Inmagic #2977
K/HS-95	Lay, A.C. and Rogers, J.G. Oak Ridge Gaseous Diffusion Plant Historical Uranium and Radionuclide Release Report. ORGDP. Martin Marietta. Inmagic #1334
Shonka 1995	Interview Notes: Tom Hanrahan. 10/95. Inmagic # 2534
SRA-96-009	Burmeister, R.E. Fitting Uranium Release Estimates of the Purge Cascade. 1996.
SRA-96-010	Burmeister, R.E. Fitting Estimates for ESA, EIVA, CIVA, and DD Pathways. 1996.

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SRA-96-011	Release Estimates for the S-50 Liquid Thermal Diffusion Plant. 1996.
US AEC	Environmental Monitoring Report United States Atomic Energy Commission Oak Ridge Facilities for the Calendar Year 1973. Inmagic # 944.
US DOE	Environmental Monitoring Report United States Department of Energy. 1977-1982. Inmagic #s 948, 949, 950, 951, 952, and 953.
US ERDA	Environmental Monitoring Report United States Energy Research and Development Administration. 1974-1976. Inmagic #s 945, 946, and 947.

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7. ELECTRONIC FILES

The following files are included on the diskette that accompanies this calculation.

<u>File Name</u>	<u>Description</u>
SRA012.DOC	This calculation in MS-WORD format
ATM10A.XLS	50%-ile Atmospheric Release Estimate for K-25 Site; EXCEL spread sheet
ATM10B.XLS	95%-ile Atmospheric Release Estimate for K-25 Site; EXCEL spread sheet
ENVIRO.XLS	Enrichment calculations for the environmental work; EXCEL spread sheet

Appendix A

SRA

August 22, 1996

Shonka Research Associates, Inc.

To: File

From: J. J. Shonka

Re: Task 6: Review of Release Fraction Literature

Memo No. JJS.048

cc T. Widner
J. Buddenbaum

A study of release fractions has been made. The release fractions under study relate to the fraction of uranium that would be released from a loss of process gas (UF_6) at a gaseous diffusion plant. Task 6 of the Oak Ridge Dose Reconstruction is charged with independently estimating the releases of uranium from the Oak Ridge Reservation, including the gaseous diffusion plant. The effort is directed towards establishing any errors in reporting that would cause significant changes in the site asserted releases. In the earlier feasibility study, the site asserted releases as documented in ORO-890 for the maximum year of releases was used as a screening estimate. Based on that estimate, the offsite impact of uranium releases was small compared to other significant contaminants of concern. Because uranium was used in large quantities at the Oak Ridge Reservation, this result seemed counter-intuitive to members of the review panel. The review panel suggested that in this phase of the project the sources of uncertainty and the potential for unmonitored releases be considered.

Many of the releases that form the basis of ORO-890 were not measured, but were asserted from accountability records of site operations. These records would assert that a release occurred at a given place and time and that a given quantity of Uranium was lost and presumed released. While not explicitly stated, either in ORO-890 or in its source documentation from the ORGDP, the previous source terms assumed that virtually all of the material was released with none of it held up on the surfaces of the plant and other systems. This would imply a large value, approaching 100% was used as a release fraction. This memorandum summarizes available scientific literature on UF_6 releases, and concurs that a 100% release fraction is possible and appropriate given the uncertainty in the size of release and other parameters.

Discussions were held with LMES technical staff concerning UF_6 behavior in a release. This behavior is qualitatively summarized as follows: for a release of UF_6 to occur, the source material must be at elevated temperatures and pressures relative to ambient conditions. At ambient temperatures, UF_6 is a solid. Under process diffusion plant conditions, the released UF_6 forms a white cloud from reactions with atmospheric moisture in air, forming hydrated uranyl fluoride ($\text{UO}_2\text{F}_2 \cdot \text{H}_2\text{O}$) and hydrofluoric acid (HF). The reactions are exothermic and, combined with the process thermal energy present inside of a gaseous diffusion plant, result in the rapid ascent of the white cloud of reaction products to the ceiling. (For ground level releases with pressures and temperatures just above the triple point, the cloud may remain near the ground.) The ceiling of cells of a gaseous diffusion plant are provided with periodic vents. The size and spacing of the vents is designed for removal of the hot air given off by process equipment, and not necessarily for removal of noxious contaminants. If the release occurs immediately below a vent, essentially all of the released material will exhaust through the vent. If the release occurs at some distance from a roof vent, the material rises to the ceiling and spreads out, gradually forming particulate (which partially deposits on surfaces) while the material migrates to and is entrained into nearby vent exhausts. In most cases, nearly all of the uranium is exhausted from the plant unless special measures are taken to limit the release (close all vents and inject steam into the cell to fully react the UF_6 and provide condensation nuclei to enhance fallout, has been attempted for example). Thus, a release fraction of 100% is appropriate and does not offer a substantial degree of conservatism, if any. Supporting evidence for this assumption is the lack of significant fallout found in several release tests of UF_6 (both indoors and outdoors) to the environment.

One of the most applicable reports is K-GD-916, "Containment of Released Uranium Hexafluoride" by R. L. Ritter (11/7/73). This report, as reviewed by the analyst, is an unclassified, redacted version of a short report of 1973 release studies in Cell K-902-5.9. Fifteen minute releases at rates from 10 to 100 grams UF_6 per minute were visually observed by a window placed into the cell wall, with air concentrations measured inside and outside the cell. Deposition studies were made using one square foot fallout pans which were placed inside and outside the cell and the release fraction to outside the building was estimated. UF_6 in moist air at room temperatures rapidly forms a white cloud. Because of elevated temperatures ($>160^\circ\text{F}$) in the cell as well as other differences (such as small release rate and high air flow rates) this white cloud was never observed either in the cell, building or outdoors. The relative humidity outdoors was likely low as well, given the time of year. Thus, heating the outdoor air to an ambient temperature of more than 160°F would also produce in-cell air at a low relative humidity. A haze was observed sometime after the larger of the releases began. The white cloud was observed when the releases were made outside of the cells, although that portion of the report remains classified. Typical release fractions to the environment were asserted to be between 20% and 40%, although one of six experiments where air concentrations were measured had a

significantly lower release fraction of 5.3% that could not be explained by the authors. If you remove the low value, the average release fraction measured was 27% +/- 7% (one sigma). The cells had background deposition rates of two to four $\mu\text{g U/sq.ft./24 hours}$. Negligible fallout rates were observed above background inside the cell for release rates of less than 50 g/min. At higher release rates, slight fallout was observed in the cell and on the roof of the building. There is a roof vent immediately above the cell where the experiments were conducted. An average of 900 linear feet per minute was observed with high variability over the 32 square feet of vent area for an assumed 28,800 CFM flow rate. On three of the runs, the louvers were in the 10% open position which caused even greater air flow variations. The measurements of concentration across the varying air flow in the vent made asserting a release fraction difficult, and the author suggested that his data was more qualitative than quantitative.

The release fractions asserted by the author were based on a dynamic experiment that was difficult to control. The release would mix into the cell volume, which could likely be modeled by a first order linear kinetics model ($\exp(-\mu t)$) which would have an exponential response (with a time constant consistent with the cell air changes per hour) to a step change in input rate (e.g. from 0 to 50 grams per minute). The cell concentration was likely not at equilibrium until the release was terminated and the residual contents of the cell equilibrated (if they ever did). The cell contents, which are time varying, would mix, perhaps imperfectly with the access tunnel ventilation flow rate and be ejected out the vent. The vents were about 25 meters above the ground, and heat and ventilation cause the plume to be ejected another 15 meters up, for an effective release height of about 50 meters. The flow rate across the ventilation system was non-uniform, the air concentrations were non-uniform, and the releases were only maintained for 15 minutes into the cell, which would be smeared out in time by the mixing into the cell volume and exhaust. Thus, the observation by the author that the release fraction data should be viewed as qualitative are appropriate. The data was not analyzed for mass balance. A primitive mass balance could be derived from the fact that even though the largest releases had between 1200 and 1500 grams released, only tens of micrograms per square foot were observed as fallout. With cell areas of substantially less than millions of square feet, fallout accounted for less than one gram of material or at most a few grams) over 24 hours. Where did 99.9% of the material go? It had to either accumulate in an undiscovered area of the process building, or it had to go out with the airflow. Thus, although the analysis emphasized the air concentration, this was a dynamic variable that was difficult to measure. When one simply considers the lack of fallout of the material, release fractions of 99% or more cannot be eliminated and are consistent with the data. The applicability to releases to moist air at lower temperatures is not entirely straightforward. Clearly, the reaction rates were suppressed due to lack of atmospheric moisture. Thus, the formation of particulate would occur at a reduced rate, perhaps suppressing the amount of uranium that would appear as plate-out. The lack of equilibrium in the measurements

would be one reason for the data to be biased to low release fractions. Additionally, the ratio of the sampler flow rate to the vent flow rate is subject to considerable uncertainty, providing another potential source of bias. Finally, there is potential for a fraction of the release to exhaust from distant vents, since the air containing the reacting UF_6 would rise to the ceiling of the cell.

Report K-D-1894, "ORGDG Container Test and Development Program Fire Tests of UF_6 -Filled Cylinders" by A. J. Mallett (1/12/66) reported the destructive testing of potential shipping containers to fire environments during October of 1965. The tests were destructive, with two cylinders and three capacities of 5, 55 and 250 pounds of UF_6 used at an enrichment of 0.22%. The tests were primarily designed to observe the cylinder behavior in a fire. Of the data taken, the lack of significant air concentrations and fallout were noted. This report provides additional justification that the behavior of plumes of UF_6 is not affected in a significant fashion by the relatively high mass of the molecule, and the plume behaves in a manner similar to other chemical fire plumes.

Report KY/L-1213, "Assessment of Consolidated UF_6 Release Studies" by D. E. Boyd, C. G. Jones, and S. F. Seltzer (9/7/83) reported the efforts by DOE to consolidate UF_6 studies at the various plants to avoid duplication of effort. The report summarizes the studies each of the plants was conducting and was primarily a source of references on the related work. Work summarized from the Paducah Gaseous Diffusion Plant's KY/L-725 report "UO₂F₂ Particle Size Analysis by the Coulter Counter Method" indicated that the measured UO₂F₂ particle size ranged from 0.8u to 40u with the predominant size in the 0.8u to 2.5u range.

Report KY-795 "Fallout of Uranium During UF_6 Releases (UU)" is a 1/6/94 report by T. J. Mayo that summarizes data from two reports written in 1975 (KY/L-694 and KY/L-765). These reports discussed experiments where a heated bulb was charged with 215 grams of UF_6 and 14 grams of SF_6 at 58 psia. The contents were allowed to escape in the field, which resulted in the release of 160 grams of UF_6 and 10 grams of SF_6 . Eight releases were characterized in the first report, four in the second report. It was necessary to perform the releases late in the day to avoid excessive atmospheric dispersion which resulted in experimental data below the detection limit of the experiments. A chemically treated filter paper was used to measure the HF and UO₂F₂ as a function of distance. Measurements were made to distances of up to 400 meters, with small quantities of uranium observed. The author argued that the observations appear to support a conclusion that one cannot assume a large fraction of the uranium will quickly fallout from a cloud of reacting UF_6 .

Between 2/25/76 and 8/17/76 a total of 57 test releases involving a total of 2032.5 grams of UF_6 were made in K-33. The need for the tests was driven by the change in the K-33 ventilation system.

The project has a cover letter concerning this experiment and is attempting to recover the document.

Tackle Found

of these tests

K/D-6092 Analysis of the June 5, 1989, UF₆ Release Test (2/93) by S. G. Bloom is an analysis of a cooperative release study performed in France. A series of UF₆ release tests were conducted by the French at their government test site at Bordeaux, France. About 150 Kg of UF₆ was released over 30 minutes at a height of 3 meters. Information included meteorological data, uranium and fluorine concentrations, particle size distribution, deposition data and visual observations. The US interest was in developing the data to benchmark an environmental transport code for UF₆ that accounted for chemical and physical transformations in a chemically reacting plume. The data showed a small particle size distribution for uranium that experienced deposition velocities from 0.01 to 2 centimeters per second, with an Andersen Impact Sampler measure mean particle size of about 3 μmeters. The data showed decreasing deposition with distance.

Finally, Report KY-L-824, "The Application of the Gaussian Plume Model Equation to UF₆ Releases" by T. J. Mayo (4/15/76) reported both SF₆ releases and studies of uranium fallout at the Paducah Gaseous Diffusion Plant. The conclusions of the report were that fallout would not be a major factor in reducing uranium concentrations in air at least to distances of several hundred meters.

Note added 11/1/96:

Earlier drafts of this memorandum were provided to staff (B. Manninen) at an operating gaseous diffusion plant (Portsmouth) for review and comment (See JJS.049). Telephone conversations were later held to obtain their comments and reactions to the assertions of release fraction. Prior to this memorandum, the staff felt that releases immediately below vents would have near total release fractions, but that would not occur for releases from equipment located some distance from a vent. In the discussions held after they had reviewed this data, their position changed to one which agreed with this memo, largely because of the particle size distribution observed in the French experiments (K/D-6092).

MEMORANDUM

Date: 10/23/96

To: Joe Shonka

From: Jack Buddenbaum

cc: Tom Widner, Jennifer Cockroft

Subject: 1986 - 1995 K-25 Uranium Airborne Releases

I have summarized below K-25 air release estimates for 1986 - 1994. The 1995 numbers can be obtained from a web site identified below. I have also attached to this memo, three FOIA incident notification summaries from the National Response Center. These reports describe K-25 uranium releases that may have not been included in Task 6 estimates. Let me know what you think.

K-25 Atmospheric Releases for 1986 - 1996

<u>Year</u>	<u>Curies</u>	<u>Kilograms</u>
1986	0.001	0.196
1987	0.0003	0.4
1988	0.0011	1.71
1989	0.0004	1.11
1990	0.0011	2.01
1991	0.0240	40.22
1992	0.0640	112.39
1993	0.01	12
1994	0.008	10
1995	*	*

* - The Oak Ridge Reservation Annual Site Environmental Report for 1995 can be retrieved from http://www.ornl.gov/Env_Rpt/asr95/asr.htm.

Let me know if you can retrieve the 1995 K-25 releases numbers from this web site. I can retrieve them here as soon as our IT coordinator returns to the office. Please cc-mail to me ASAP the updated spreadsheet that includes 1944 - 1995 atmospheric releases for K-25.

Let me know if you have questions or comments.

Thanks,

Jack

NATIONAL RESPONSE CENTER

*** FOIA INCIDENT REPORT 7179 ***
FOR 1985

INCIDENT DESCRIPTION

Report taken by BROWN on 31 at 1614
Incident type: F
Affected Medium: ATMOS
The incident occurred on 30 JUL85 at 1730 local time.
Weather: N Sea: N Current: N
Color: N Wind: N
Sheen Size: N
Vessel/Vehicle:
Consignee:

SOURCE/CAUSE OF INCIDENT
RESIDUAL IN LINE BLOWN OUT STACK

INCIDENT LOCATION

OAKRIDGE GASIOUS DIFFUSION PLANT BLDG K-31 HWY 58 OADRIDGE
TN ROAN CNTY

RELEASED MATERIAL(S)

CHRIS Code: MIS URANIUM HEXAFLORIDE (GAS) RAD
Qty Released: 2.2 LBS
Qty in Water: N

DAMAGE

Injuries: 0 Fatalities: 0
Damages:

REMEDIAL ACTIONS

NONE
]

REPORTING PARTY

Organization: DOE
Address: BX E OAKRIDGE TN 37831
State: TN

Calling for Responsible Party: Y

NOTIFICATIONS

B

EPA AND STATE OF TN

ADDITIONAL INFORMATION

*** END FOIA INCIDENT REPORT 7179 ***

NATIONAL RESPONSE CENTER

*** FOIA INCIDENT REPORT 16444 ***
FOR 1988

INCIDENT DESCRIPTION

Report taken by RCP on 25-SEP-88 at 1646

Incident type: F

The incident occurred on 25-SEP-88 at 1525 local time.

Vessel/Vehicle:

SOURCE/CAUSE OF INCIDENT

SPILL FROM A MIXING TRUCK. DUE TO OPERATOR ERROR.
TRUCK WAS PARKED AT THE FACILITY

INCIDENT LOCATION

NEAR HWY 95

County: ROAN

City: OAKRIDGE

St: TN

RELEASED MATERIAL(S)

Chris

Code:	Material Name:	Total Qty:	Units:	In Water:	Units:
RAM	URANIUM/F006	10.00	LBS	0.00	NON
	0.00	0.00			
	0.00	0.00			

DAMAGE

Injuries:

Fatalities:

Evacuations: 0

Damages: 0

Amount:

REMEDIAL ACTIONS

MATERIAL WAS PICKED UP AND PUT BACK INTO THE MIXER
CLEANED.

REPORTING PARTY

Organization: DEPT OF ENERGY OAKRIDGE GASEOUS

Addr:

State: TN Zip: 37831-

Calling for Responsible Party: 1

SUSPECTED RESPONSIBLE PARTY

Organization:

Addr:

State: Zip:

NOTIFICATIONS

EPA Region: 4 Time: 1703
MSO/COTP: Time: ?
Caller Notified: TN EMER MGT
Others Notified: NRC-1702, DOE-1707

ADDITIONAL INFORMATION

*** END FOIA INCIDENT REPORT 16444 ***

NATIONAL RESPONSE CENTER

*** FOIA INCIDENT REPORT 16284 ***
FOR 1988

INCIDENT DESCRIPTION

Report taken by AKL on 21-SEP-88 at 2356
Incident type: F
The incident occurred on 21-SEP-88 at 2150 local time.
Vessel/Vehicle:

SOURCE/CAUSE OF INCIDENT
LARGE DRUM/ FELL OFF PALLET AND BROKE

INCIDENT LOCATION
OAKRIDGE GASEOUS DIFFUSION PLANT

County: ROANE City: OAKRIDGE St: TN

RELEASED MATERIAL(S)

Chris
Code: Material Name: Total Qty: Units: In Water: Units:
NCC URANIUM/F006 1500.00 LBS 0.00 NON
0.00 0.00
0.00 0.00

DAMAGE

Injuries: Fatalities: Evacuations: 0
Damages: 0 Amount:

REMEDIAL ACTIONS

SPILL HAS BEEN CONTAINED AND IS BEING CLEANED UP,

REPORTING PARTY

Organization: DEPT OF ENERGY, OAKRIDGE, TN
Addr: FEDERAL OFFICE BUILDING
State: TN Zip: 37831-

Calling for Responsible Party: 1

SUSPECTED RESPONSIBLE PARTY

Organization:
Addr:
State: Zip:

NOTIFICATIONS

EPA Region: 4 Time: 24
MSO/COTP: Time: ?
Caller Notified: TN EM
Others Notified:

ADDITIONAL INFORMATION

*** END FOIA INCIDENT REPORT 16284 ***